Factors affecting productivity and quality in the Extrusion of Polyvinylchloride Powder

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SUMMARY

Microscope examination of material from a single screw extruder screw channel following removal from the extruder barrel showed that the fusion mechanism and fusion rate depended on the lubricants used. One of two different fusion mechanisms occurred depending on the formulation. The "rear of the channel fusion model" produced rapid fusion and promoted mixing, while the "forward fusion model" resulted in incomplete fusion and the presence of unfused PVC particles in the extrudate.

Rate of fusion depended on the level of the various additives. Thus increasing the level of incompatible lubricant decreased the fusion rate, while incorporation of process aids, impact modifiers and filler increased the fusion rate. Output rate was dependent on the rate of fusion with the result that increasing the fusion rate decreased the output rate. This means that factors causing fusion rate variations will also result in output rate fluctuations.

For maximum thermal stability an optimum fusion rate exists as earlier fusion results in overheating while late or incomplete fusion results in late or inadequate incorporation of stabiliser.

The existance of two possible mechanisms explains why, following (for economic reasons) the superseding of granule extrusion by direct powder extrusion, British extrusion practice favoured the imported twin screw extruder, whereas United States practice was to continue with the cheaper rugged single screw machine.

Preface

In common with many other plastics processes, the techniques used by the plastics extrusion industry have advanced at a much faster rate than the understanding of the basic technology. The disadvantages of this situation become apparent when one considers the high scrap rates that can occur when "trial and error" methods are used to solve production problems. However, it must be admitted that many answers have been found this way even though the reasons for them have often not been understood.

For this reason it was decided to present this technological investigation against a background of industrial practice. The experimental work has been arranged in two sections (covering the two separate but inter-related stages of mixing and extrusion) to follow data obtained from factory surveys as well as published literature and then to bring them together in the discussion. It is intended that, by using this form of presentation, the factors investigated will be presented in a useful and practical way.

A "Glossary of terms" has been included at the end as some of the terms used are part of the "jargon" peculiar to this industry. Although in many cases no precise definition exists, they nevertheless often enable simpler clearer descriptions to be made than would otherwise be possible.

Factors affecting productivity and quality in the Extrusion of Polyvinylchloride powder

by

G. M. Gale

Factors affecting productivity and quality in the extrusion of polyvinyl chloride from powder

1. Introduction

In the extrusion of unplasticised PVC, the limits within which an extruder can work are very narrow compared with most other thermoplastics. Assuming the machinery variables to be fixed, any slight change in material variables could easily move the process outside those limits, resulting in, at best, rejected product, and, at worst, shut down due to "catastrophic" degradation.

From discussions with most of the larger PVC extrusion companies in the UK and one very large Dutch company, it appeared that the changeover from granule to direct powder extrusion introduced extra material variables and at the same time imposed even narrower operating limits. At the time of the discussions, a few of the companies visited were extruding directly from powder, one was buying in granules from a polymer manufacturer, one was buying-in premixed powder and the rest were in plant compounding using compounding extruders to convert premixed powder to granules. The few who were extruding directly from powder had had to overcome various problems, while others had carried out trials but were progressing very slowly owing to various problems encountered. The problems were not clearly identified but were manifested as unacceptable quality, uneconomic output rates and physical property or dimensional variations. There was considerable interest in the possible economic savings that might be obtained.

A general picture was formed of the requirements of the problems but the basic difficulty was to determine more precisely what information was required and in what way experimental work should be carried out to determine this. It was recognised that the answers to this would have, in part, to be found from industrial sources. Following the discussions with various firms a critical review of the literature was made to determine what information was already available and this is reproduced in condensed form. A study was made of results obtained by Daniels and Matthews (1.) in interfirm comparisons of companies producing PVC pipe. Their survey was used for a comparison of efficiency, wastage, scrap, manpower, etc., on an interfirm basis. In this study the results have been used for a comparison of extruders grouped according to type and determining the areas where deficiencies occur.

The basic steps of the process are illustrated in figure 1. Each step may be represented by one or more operations either by many machines or several operations carried out within one machine. The first step of premixing the additives with a high speed mixer was examined for effects of variables in this process. The step of direct conversion from powder compound to product with one extruder was then examined in detail using a single screw extruder. The two stages have not been divorced; the second step being used as an experimental tool to examine the first. Variables in mixing are, in the end, only important if they can be associated with variables in extrusion.

The problems associated specifically with the methods alternative to direct powder extrusion i.e. internal mixer compounding, extruder compounding, granulating as well as two stage extrusion and tapered screw extrusion are outside the scope of this work. These processes, and the developments leading up to present day powder extrusion techniques are described as they are significant in appraising the problems that have appeared.

In considering variables, this work has not been confined to batch-to-batch variations but has concentrated on the effects of controlling the behaviour of the compounds by attention to lubrication. Allowances have to be made for different extruder requirements. The inter-firm survey showed that it is normal for a factory to have

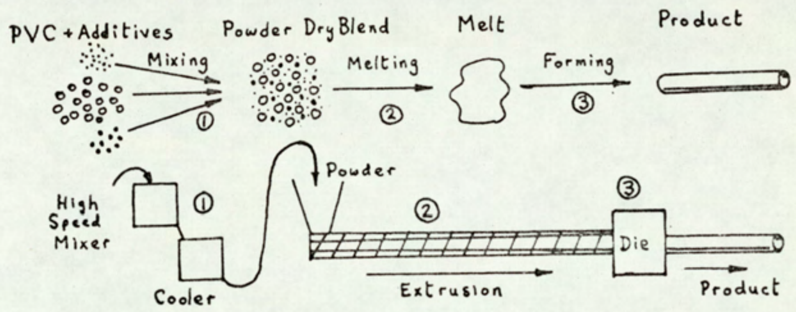


Figure 1. Basic steps in PVC powder extrusion.

a single mixing plant which has to supply a range of extruders of different types sizes and ages fitted with dies of various sizes.

The problem was therefore to determine which of the variables were the most important, why they were the most important and how they should be controlled. In order to do this it appeared that more about the mechanisms of PVC powder processing needed to be understood, so that the requirements could be recognised.

1.1 The PVC extrusion process

In order to understand the special requirements of unplasticised PVC extrusion it is necessary to consider the overall process and the various steps that can be used to achieve this. PVC polymer is used in powder form produced by emulsion, suspension or bulk polymerisation processes, although at present suspension polymers with a particle size range 50-200 microns are mainly used in rigid PVC extrusion. The polymer in powder form has to be compounded with various additives to provide a material suitable for conversion to a final product. As PVC is thermally unstable, stabilisers have to be incorporated otherwise the PVC would rapidly decompose liberating hydrogen chloride. The stabilisers can be lead salts, Cadmium, Barium, Zinc soaps and complexes and organo tin compounds. The mechanism of degradation and stabilisation is not fully understood but lead salts such as tribasic lead sulphate appear to act as absorbers of HCl which would otherwise catalyse the decomposition reaction. Lead compounds are normally used for stabilisation of pipes and other opaque building products in the UK as they are the most effective in relation to their cost. Because of this situation this investigation has been confined to lead stabilised materials.

Lubricants are essential in PVC processing to reduce the development of heat by intensive shearing effects, to prevent material stagnation in the die as a result of adhesion to metal and to give an acceptable surface finish to the extrudate. To achieve this they are also required to have a compatibility sufficiently low to produce surface lubrication, but have sufficient affinity to the polymer to prevent an unsightly bloom. Typical materials are metal soaps and fatty acid esters. Often a mixture of lubricants is necessary to produce the required properties to the compound. Other additives that can be used are pigments, antioxidants, process aids, impact modifiers and fillers. Each ingredient places certain requirements

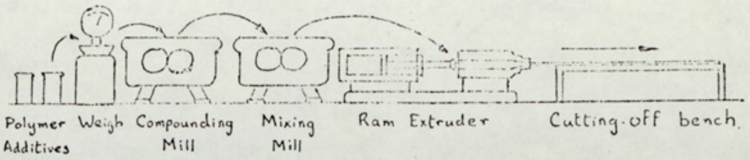


Figure 2. The ram extrusion process.

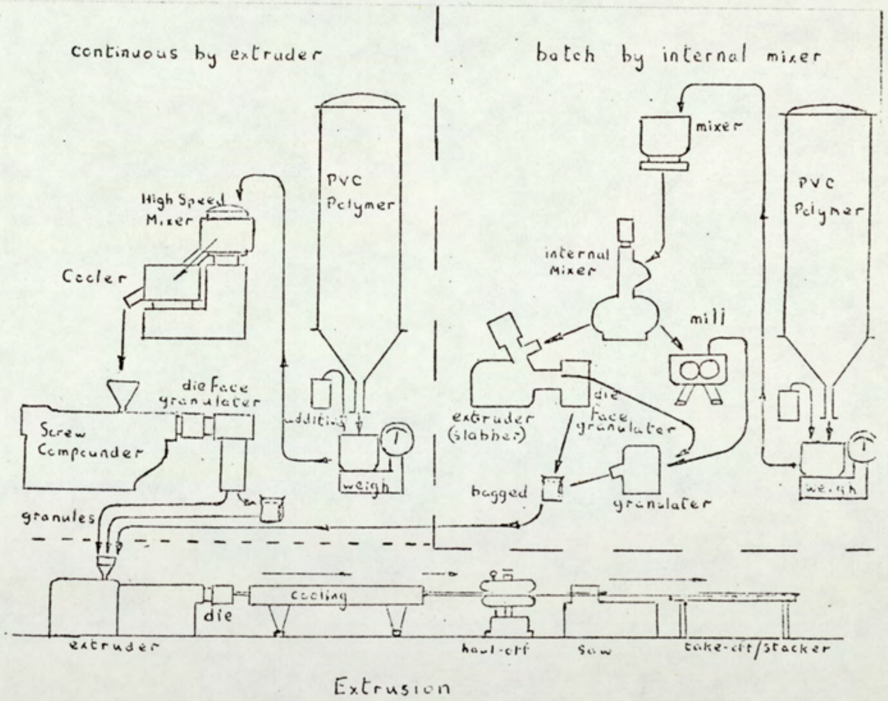


Figure 3 - Compounding stages in the production of PVC pipe

on the process and the interaction of their effects on processing will be seen to prevent their treatment in isolation. In addition the final properties and service behaviour can depend on the effects of additives on processing.

In general the overall process involves three basic stages (see Figure 1).

Stage (1) Mixing and dispersion of additives with the polymer.
Stage (2) Compounding the mix to give a homogeneous melt.
Stage (3) Forming the melt into a product.

Various combinations of machinery can be used to cover these stages. Unplasticised PVC extrusion first became commercially important during the late 1930s in Germany for production of acid resistant pipes for use in chemical plants. The processing equipment was basically adapted from rubber and cellulosic plastics which was a logical development when considering that in common with these materials PVC requires compounding with various additives to make it processable. The technique used was a simple one which avoided many of the problems associated with present day processes (see Figure 2). The PVC was compounded on the first mill, mixing continued on a second mill which also acted as a reservoir and hot "pigs" were loaded into a ram extruder. Advantages of this method were that two roll mills would adequately melt the polymer and mix in the additives so that this stage was fairly independent of the additives and did not suffer from problems of shear heat generation. Ram extruders also avoided problems of high melt viscosities and heat generation. Although output rates were relatively high, labour content was high, the extruders needed frequent cleaning and take-off and sizing was difficult to control. (2)

Plasticised PVC was initially processed on plants previously used for rubber and so that unplasticised PVC was also initially processed in this way (Figure 3). In this case we have cold mixing of the powders, followed by conversion to a melt by an internal mixer, the melt being discharged and converted by various routes to granules. This process is normally used by polymer manufacturers who require versatility for producing a wide range of compounds. The compounds are then transported to the converter who extrudes the granules in a similar manner to other thermoplastics. Advantages of internal mixing are that the compounding conditions of time and temperature can be varied to suit the formulation to ensure full gelation of the PVC and complete dispersion of all the additives. It can also handled premixes which are sticky or cohesive.

Where only one material of one colour is used and the material is free flowing, an extruder compounder fitted with a 'die face cutter' for granule production is a more economical method. It has a lower capital cost and fits in with fully automatic bulk handling, weighing, conveying and storage. As manufacturers of specialised items, such as pipes and sheets, grew in size and their products often had the same colour, the incentive was to instal in-plant compounding equipment and screw compounders were the more attractive. The introduction of the high speed mixer enabled powder mixes to be produced which had increased bulk density, improved powder flow properties, good dispersion of additives and low moisture content. This made the screw compounder acceptable for a wider range of materials than would otherwise have been possible.

In many cases the final extruder was a twin screw machine fed with granules produced by a twin screw extruder compounder. The similarity between the two machines resulted in attempts to omit the compounding extruder and feed the mixed powder directly into the final extruder (figure 4). This direct powder extrusion was considered to have advantages of lower capital costs, lower overheads and

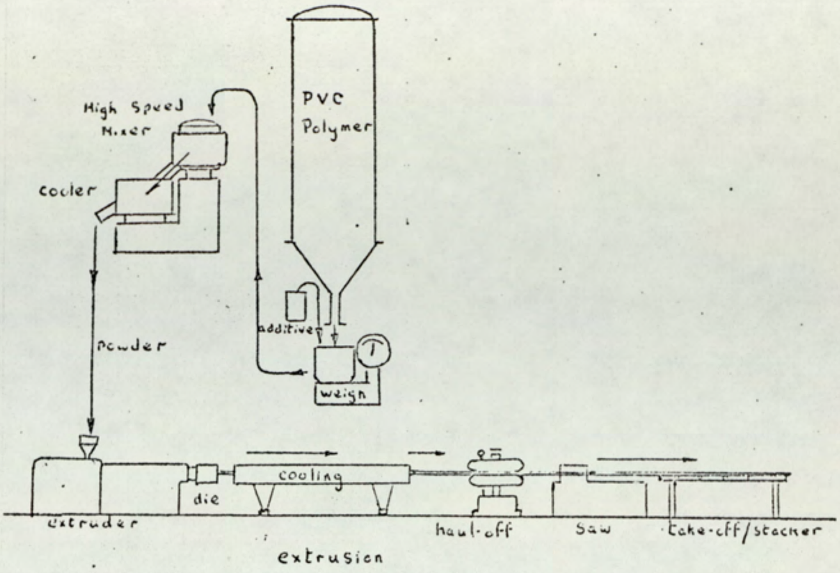
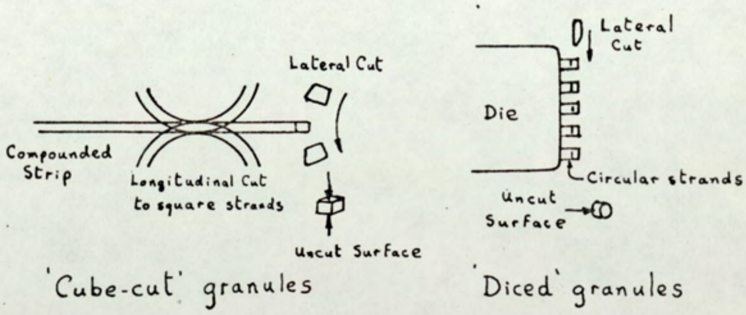


Figure 4. The direct powder extrusion process.



'Cube-cut' granules

'Diced' granules

Figure 5. Formation of granules.

reduced handling but they were often offset by reduced output rates and products having physical properties adequate only for certain applications. Direct powder extrusion obviously increases the demands made on the extruder as in this process it has to combine the function of compounding powder to a melt with conversion of the melt to a profile.

The following problems are typical of those experienced with direct powder feed extrusion -

1. Streakiness
2. Blisters
3. Rough surface
4. Separation of insoluble additives
5. Low output rate
6. Variable output rate
7. Poor heat stability
8. Poor physical properties

At this point we can consider the possible differences between powder and granule extrusion.

1.2 Differences in properties between granules and powder (see table 1)

(a) Granules

An analysis of any part of a granule should be the same as that of the finished extrusion. Uncut faces will have different surface properties to cut faces as the former will be smoother and have a higher concentration of lubricant (see diagram 4).

With granule extrusion the choice of additives such as lubricants will depend mainly on the requirements of melt properties in the die and metering zone. These will be that the melt must not overheat due to shear, must slide through the die without sticking, must weld together satisfactorily following the mandrel support in a tube die and have an acceptable product finish. It is therefore expected that additives will have little effect on feeding and melting

TABLE 1

Comparison of powder with granules

Property	Granules	Powder	Comments
Size	3-4 mm cubes discs 2-4 mm dia x 1-2 mm thick.	Irregular particles sometimes approaching spherical. Diameter usually 100-200μ.	Bulk flow of powders can be adversely effected by interparticulate forces causing bridging, ratholing etc. in hopper.
Size distribution	Usually a consistent granule size	Normal distribution usually kept as narrow as possible. Range	Segregation of particle sizes can result in varying feed rates.
Ratio of particle size to screw channel depth	1:2-1:4	1:30-1:60	Granules at a disadvantage in feeding very shallow screws. With powders, interparticulate shear can cause breakdown of conveying.
bulk density	550-750 g/l	450-620 g/l	Output rate is normally partly dependant on bulk density and therefore likely to be lower with powder. Low bulk density and restriction to air flow can result in air entrapment with powder. Fluidisation of powder can impair conveying.

and that air should have adequate passages for escape. The hopper, feed zone and to some extent the compression zone effects should be mainly a function of granule geometry and polymer properties, and in many respects will differ little in behaviour from granule extrusion of other thermoplastics.

(b) Powder

When a powder is used as a feed material, the usual properties associated with powders in general have to be considered. In addition to the requirements described above for granules one must consider the properties associated with the following.

(1) Bulk solids flow of powders in hoppers.
(2) Conveying of powders by single screw extruders.
(3) Compaction and air removal.
(4) Melting.
(5) Dispersion of additives.

Not only is the particle size much smaller than the granule, the additives will in most cases not be evenly distributed throughout the particle but will be in the form of a coating on the surface of the particle. This means that the surface characteristics of the particle will be dependent on the additives used, particularly the lubricants. Therefore in selecting additives, there is in addition to the requirements of the melt stages a requirement that these additives should also fulfil the requirements of powder flow in the powder flow in the hopper and screw conveying properties. It is therefore possible that the requirements of one stage cannot fulfill the requirements of another, or that the degree of overlap of the requirement is as narrow that extrusion conditions become very critical

1.3 Types of extruder

The requirements are expected to differ with the type of extruder used. The pattern of development has differed between USA and Europe. In the USA, single screw extruders are commonly used for rigid PVC extrusion. In common with extruder developments for other thermoplastics, screw lengths have also been increased for PVC. Additional modifications are two stage screws with venting and vacuum compaction hoppers to overcome problems of feeding powders.

European development has been to use twin screw extruders where the flights of one screw intermesh with the channel of the other in a figure-of-eight section barrel. This gives positive conveying of powder and control of compaction makes it possible to compact and sinter the material before melting commences and so reduce risks of air entrapment. Later developments have been to use venting and vacuum hopper compactors in a similar manner to single screw development. Whereas single screw extruders are essentially of similar design, twin screw machines vary considerably. The screws can be co-rotating or contra-rotating, parallel or tapered, with length ranging from $6\frac{1}{2}$ - 20 times the diameter. Compression can be achieved by varying the helix angle, channel depth, or flight land width (3). The main disadvantages are lower output rate in relation to capital cost, and limitations in bearing diameter. Other problems include the design of dies to fit the figure-of-eight barrel section and pulsations in the die arising from the pumping action of the intermeshing screws (4).

A survey described below showed that 34% of extruders used for UPVC pipe production at that time were of the single screw type, but during subsequent years the move to direct powder extrusion has been accompanied by a complete change to twin screw machines.

In view of the possible advantages of the single screw extruder and in particular the far greater UK capacity for producing this type of extruder as opposed to the twin, it appeared that a better understanding of the behaviour of powder in a single extruder was required. In particular, reconciling lack of success of single screw extruders for this application and the undesirable melting pattern shown by Menges and Klenk (5, 6) with the apparent success of single scre extruders in the USA (7) was considered necessary.

2. Survey of extrusion in the U.K.

An interfirm study of production of rigid PVC and polyethylene pipes covering six of the major pipe producers in this country was made during 1965 and 1966. This survey was presented by Daniels and Mathews (1.) as a comparison of efficiency in manpower, production of scrap material, waste, loss of production time, labour utilisation in compounding etc. In this section the results, from which the comparison was made, have been grouped together regardless of company and compared on a basis of extruder type and size in order to try and identify the main areas where technical problems exist.

The output rates for the study periods of about four weeks gave an annual tonnage for the six firms representing 60-70 per cent of the annual production of rigid PVC pipe in the United Kingdom (total based on BPF pipe group figures). This represents a relatively large sample of extrusion lines. At that time the majority of extruders surveyed were running on PVC in granule form while none of the single screw extruders surveyed were running on powder feed. The compounds in granule form were either "bought out" or "in-plant-compounded" by a variety of basically similar methods.

During the intervening years there has been a slow but steady transition from granule feed to powder feed such that the situation has changed to one where the number of extruders running on powder are now majority while the number of single screw extruders used for rigid PVC extrusion is relatively small. For the extrusion of rigid PVC it is generally assumed that the U.K. industry has always preferred twin-screw extruders to single-screw extruders and that the majority of the twin screw machines are imported from Europe. It is also usually assumed that the few single screw extruders used were supplied by UK companies who had a territorial advantage and could often supplement their own developments with those of their United States Associates. In fact this was not the case during the survey period of 1965-66. Of the 59 extruders surveyed, 20 were single screw (about one-third). Of the 39 twin screw extruders, 19 were imported machines (about one half), while of the 20 single screw extruders in use 14 were imported, (about three quarters).

The transition from granule to powder feed has changed the demands on the extruder and to meet them it appears that the twin screw machine has in general been preferred.

However, some recent unpublished surveys show that in many cases, scrap rates have increased since the transition to powder extrusion. This may be due to the narrower limits imposed by powders, but the increase in pipe sizes and the demand for higher output rates may also be significant.

The survey covered 59 extruders of which 17 were co-rotating twin screw machines, 22 were contra-rotating twin screw machines and 20 were single screw extruders. This sample provided an opportunity of comparing the relative performances of single-screw and twin-screw extruders. For most of the comparisons the powder fed extruders have been included with those fed with granules. No discrimination is made between "in-plant" and "outside" compounded materials. Although many comparisons can be made from data recorded in the factory by a survey team during a week's survey, for some comparisons data are required over a much longer period, and here reliance must be placed on factory records. The problem here is that there are interfirm differences in record keeping, while in some cases the recorded data are not specific enough. An example is the recording of non-productive time without specifying whether the line was idle from a breakdown, lack of an operator, or lack of orders. Breakdown time may not specify whether the breakdown was due to the extruder or the ancillaries.

'Percentage good extrusion' is defined as the proportion of saleable pipe made while the extruder is producing pipe, but does not include production of waste material. The causes of unsaleable pipe can be poor physical properties, poor appearance or dimensions outside specified tolerances. The first two properties are usually a function of extruder and die while the pipe dimensions can equally be dependent on the performance of the haul-off. All these deficiencies can of course be caused by variations in raw materials. Changes in physical properties such as bulk density and flowability in the feed zone will effect pipe dimensions. 'Scrap' is defined as unacceptable pipe which can be granulated and re-extruded, while 'waste' is non-extrudable and usually the result of thermal decomposition. For the purpose of comparison the performance of the extruder is taken as the performance of the line, as it is assumed that the average performance of the ancillaries will be the same for both types of extruder.

It is common practice in calculating output rates of different size extruders to use the square of the diameter as a 'scaling-up factor'. Similarly, the performance of extruders of different sizes can be compared by dividing the output rate by the square of the screw diameter. With the twin screw machines, comparisons are made more difficult by the wide range of output rates obtained from a number of extruders of the same screw diameter.

Figures 6, 7 and 8 give plots of 'percentage good extrusion' against' output rate divided by the square of the diameter' for single screw, co-rotating twin screw, and contra-rotating twin screw extruders respectively. The overall performance of the three types in terms of percentage good extrusion can be more

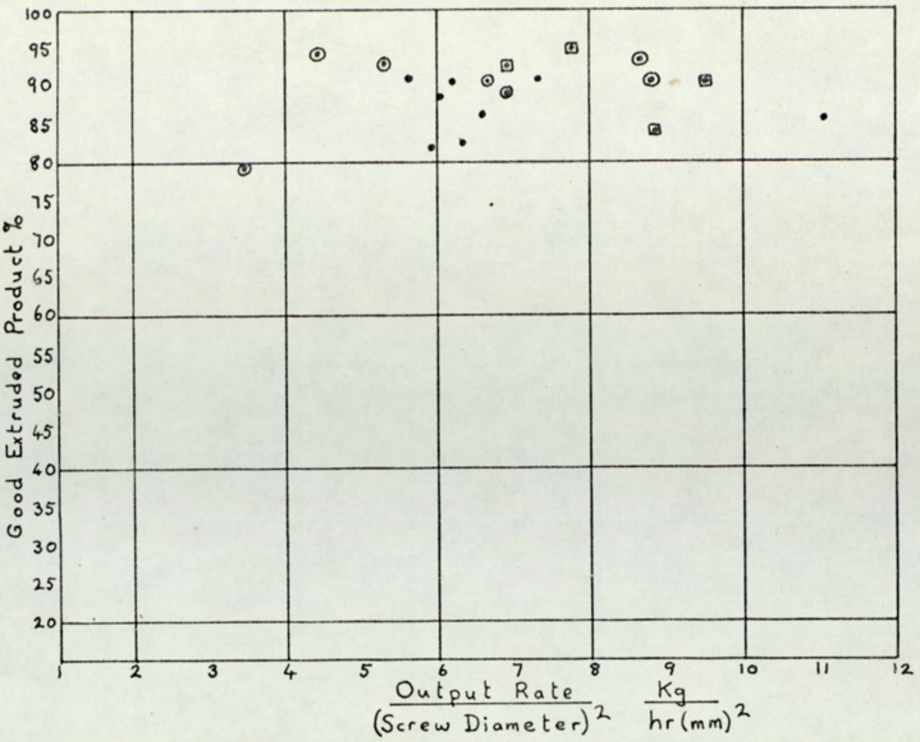


Figure 6. Performance in terms of good extruded product: single screw extruders.

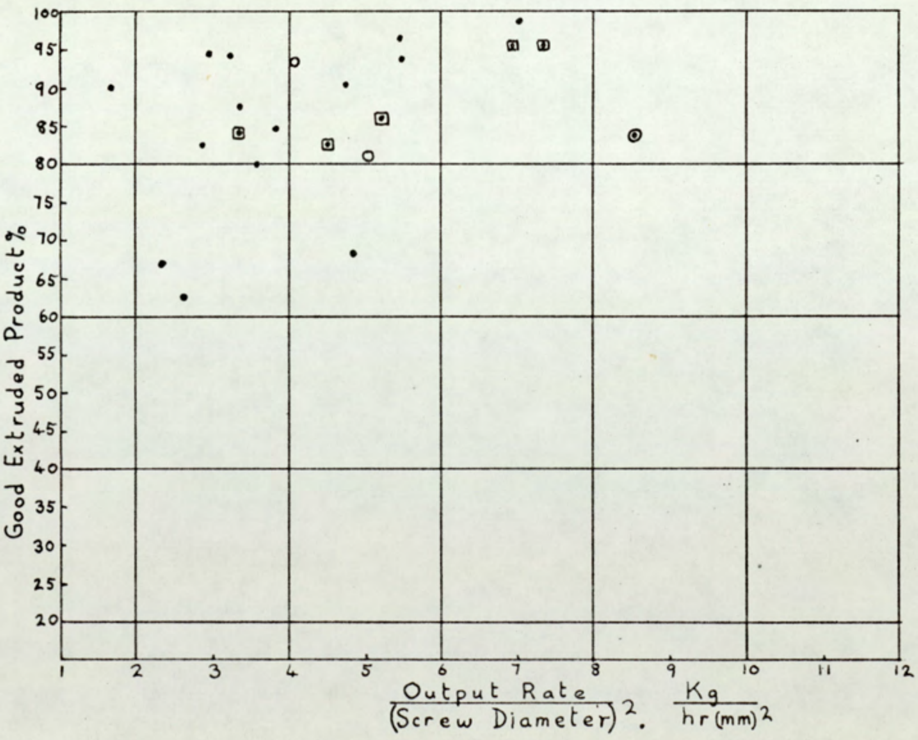


Figure 7. Performance in terms of good extruded product:- co-rotating twin screw extruders

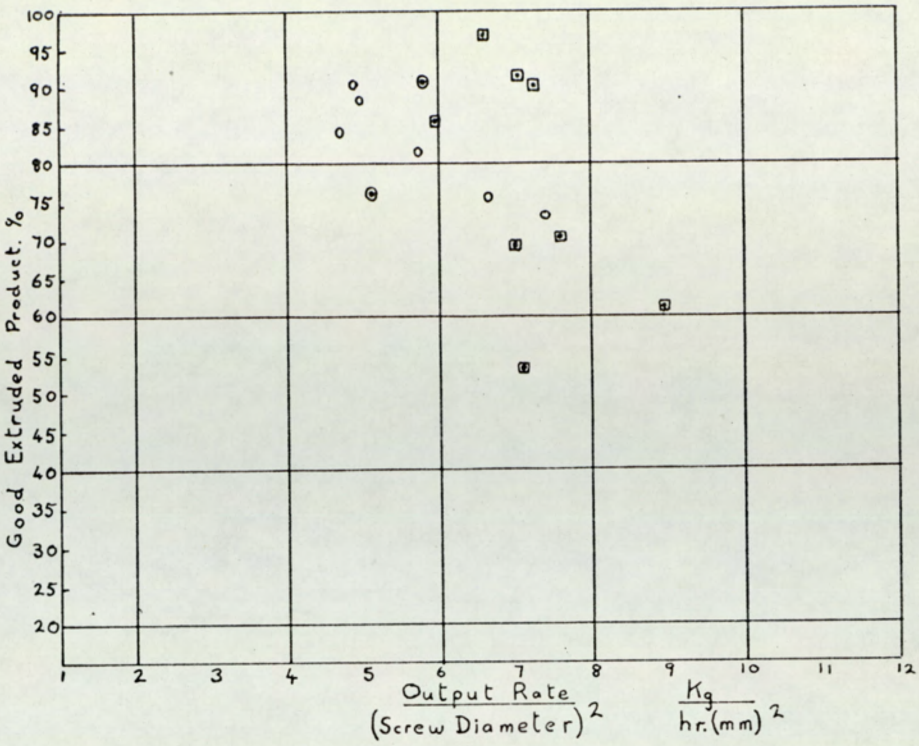


Figure 8. Performance in terms of good extruded product:- Contra-rotating twin screw extruders.

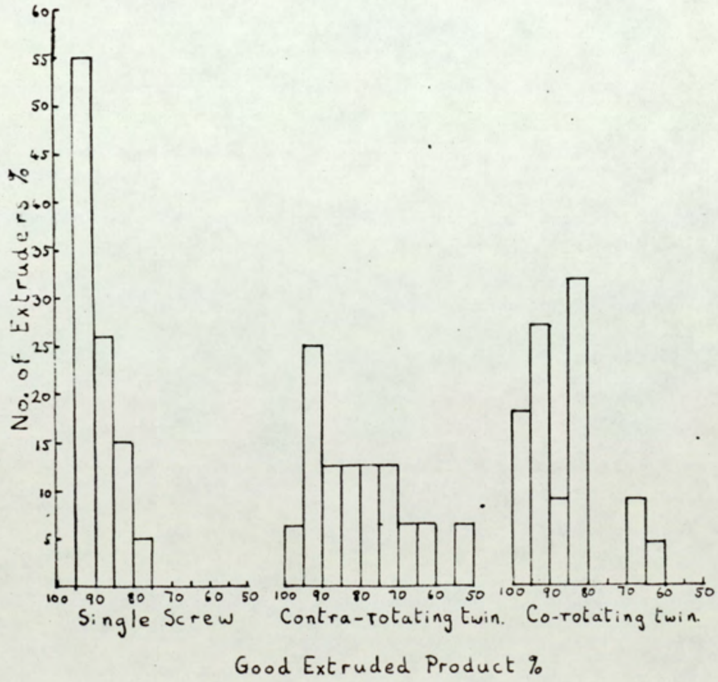


Figure 9. Comparison of extruder types in terms of good extruded product.

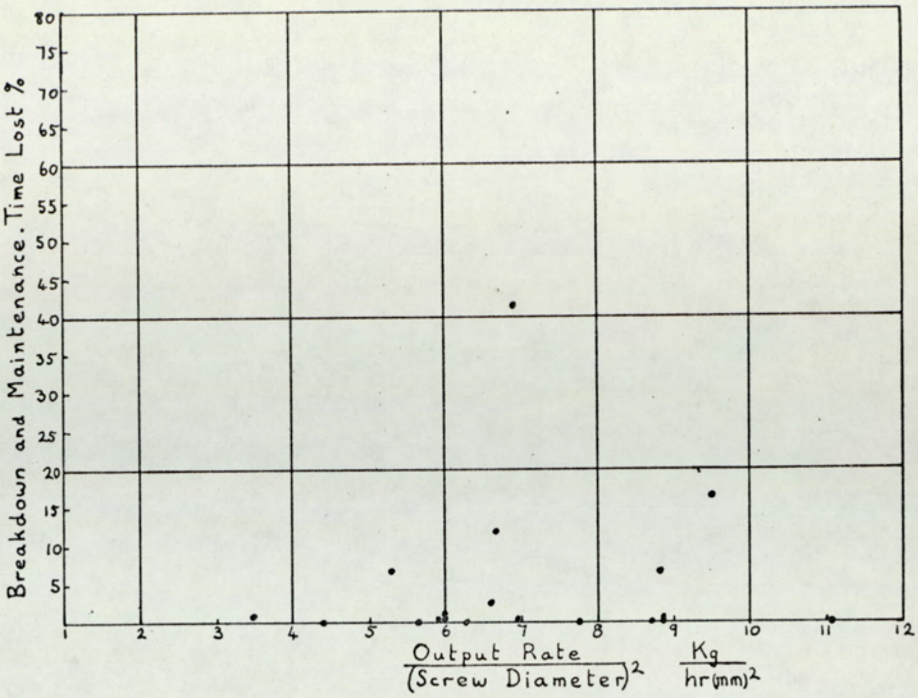


Figure 10. Time lost due to breakdown and maintenance:- single screw extruders.

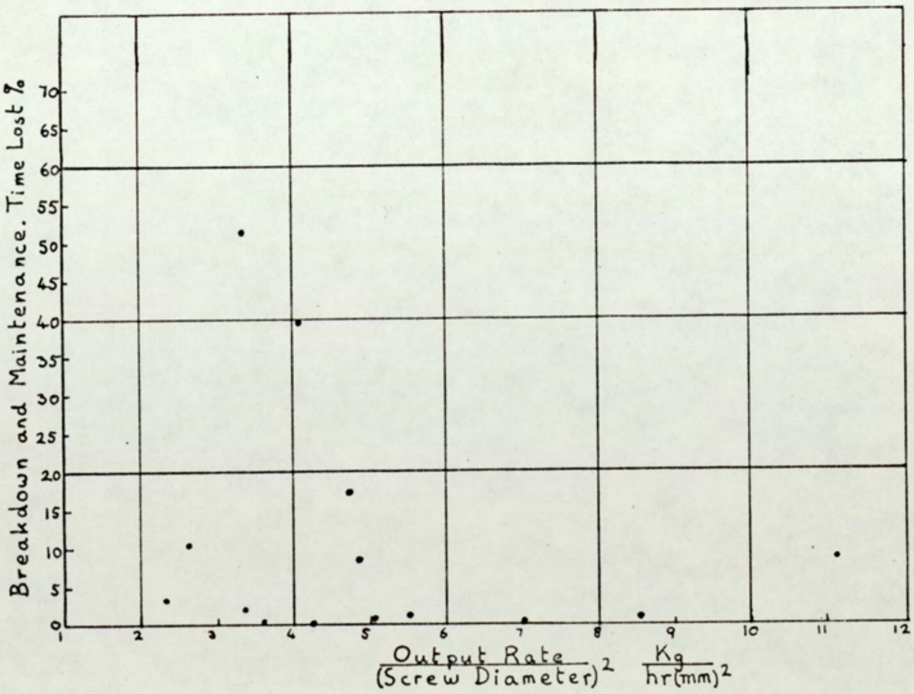


Figure 11. Time lost due to breakdown and maintenance:- co-rotating twin screw extruders.

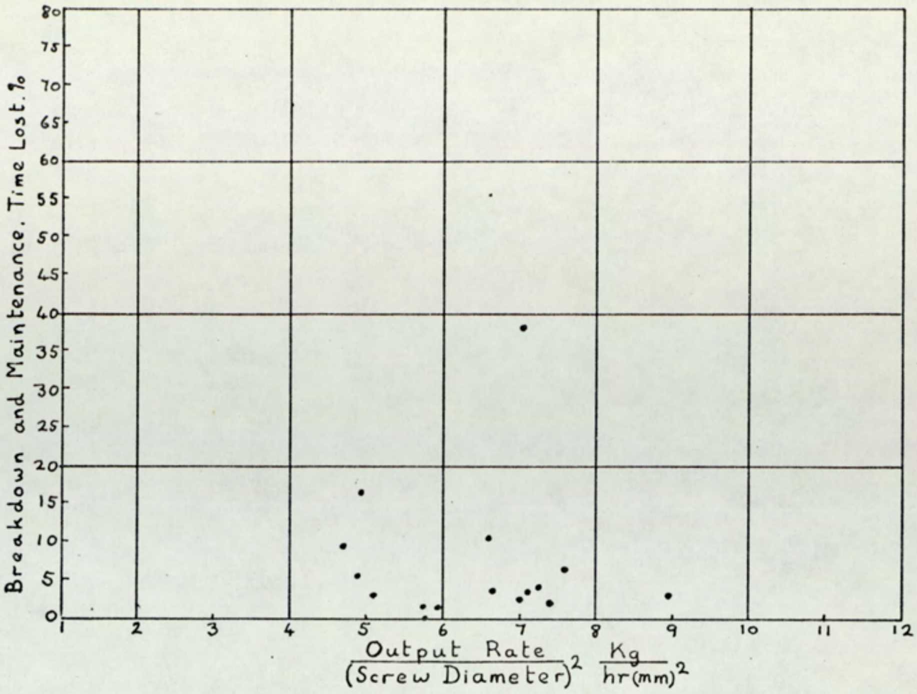


Figure 12. Time lost due to breakdown and maintenance:- contra-rotating twin screw extruders.

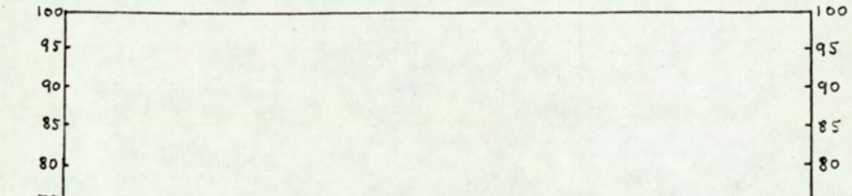


Figure 13. Comparison of extruder types in terms of time lost due to breakdown and maintenance.

easily compared in figure 9 which shows the relative numbers of extruders in bands of 5%.

For single screw extruders there appeared to be no correlation between percentage good product and either output performance or pipe size. A similar situation appears to occur for co-rotating twin screw machines although three machines gave poor performances in both output and percentage good product. With contra-rotating twin screw extruders, the percentage good product did appear to fall with higher output rates. The output rates in relation to screw diameter were generally higher for the contra-rotating extruders than for the co-rotating machines.

The overall comparisons indicate that the lines using single screw extruders were as good as or even better than those using twin screw extruders. A similar comparison restricted to machines in factories employing both twin screw and single screw extruders gave a very similar result. At least one of the very poor performances was observed to have been caused by "haul-off" problems.

Figures 10, 11 & 12 show plots of 'percentage time lost due to breakdown' against 'output rate divided by the square of the screw diameter' for the three types of extruders. Time taken by planned routine maintenance has been included in breakdown time. These data were not available for all the extrusion lines in the survey. Comparison of the figures shows approximately equal performance between lines with the three types of extruder. For all three types one or more points fall well outside the main group, but at least one of these was due to a major ancillary breakdown.

In the comparison of 'time lost due to thermal decomposition', the performance will be a function of extruder and die, and less dependent on ancillaries than in the comparisons of 'percentage good

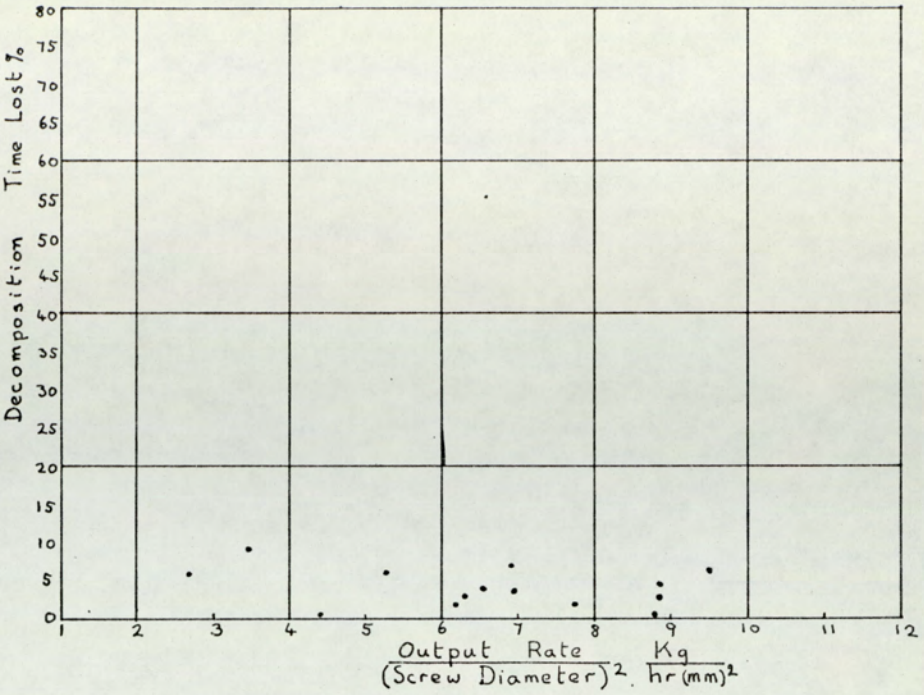


Figure 14. Time lost due to thermal decomposition:- Single screw extruders.

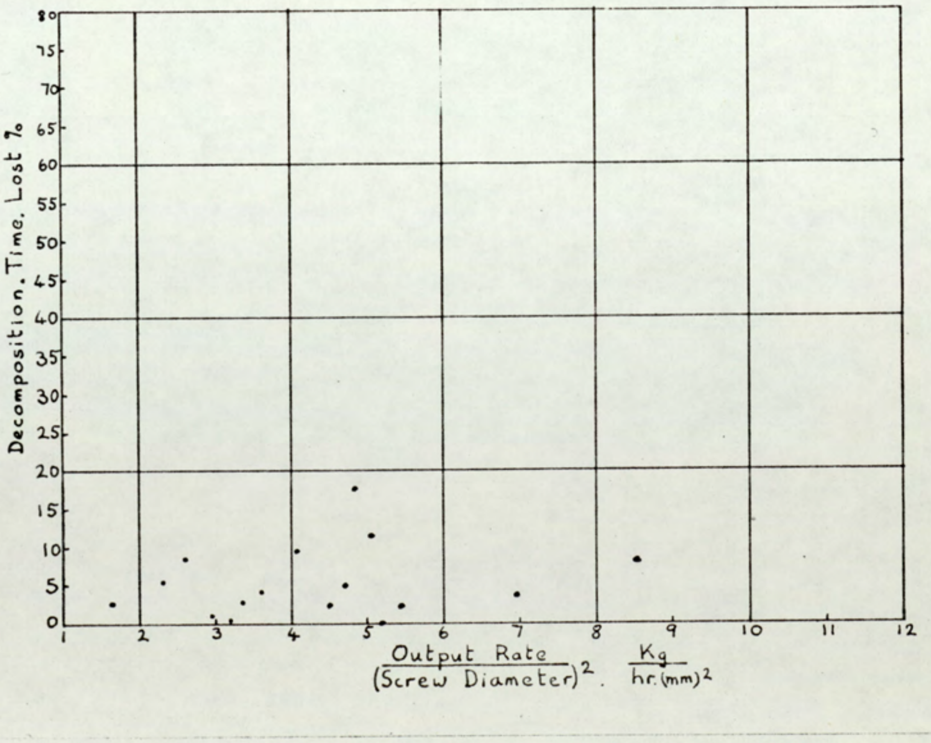


Figure 15. Time lost due to thermal decomposition:- co-rotating twin screw extruders.

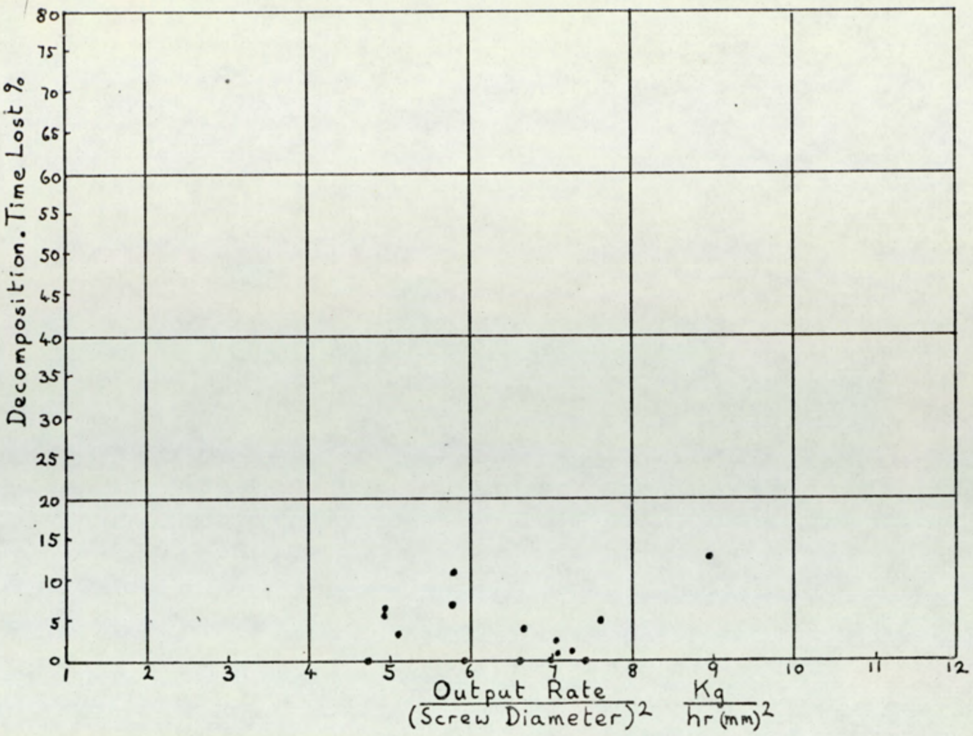


Figure 16. Time lost due to thermal decomposition:- Contra-rotating twin screw extruders.

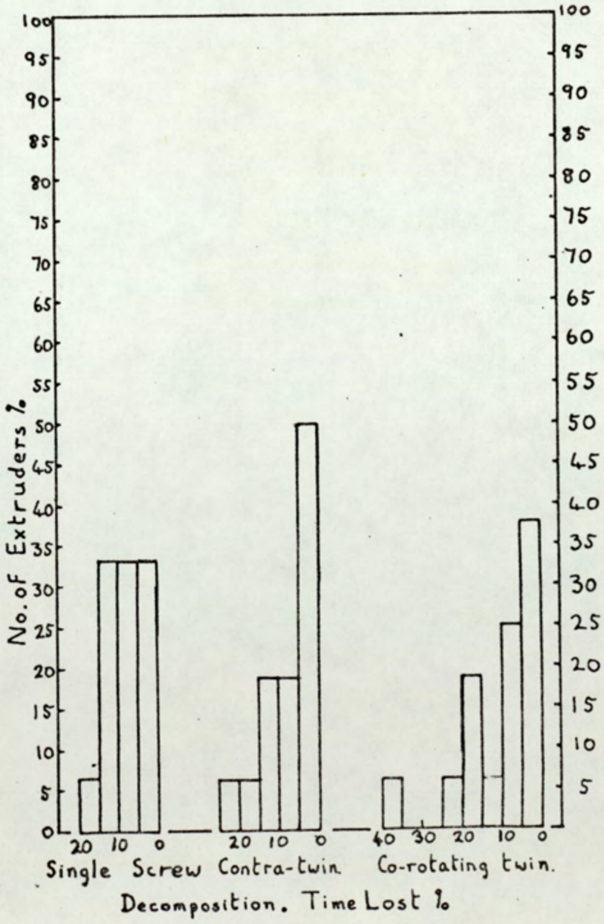


Figure 17. Comparison of extruder types in terms of time lost due to thermal decomposition.

product’ and breakdown time. The few particularly poor results occurred within the same factory and may have been a function of temperature control or thermal stability of the compound. From figs. 14-17 there appears to be no relationship between thermal decomposition problems and any of the factors of extruder type, output in relation to screw size or pipe size. The co-rotating twin screw extruders were overall marginally inferior to the two other types.

Overall output rate was plotted for the three extruder types against screw diameter. For the twin screw extruders, most of the machines were represented by two screw diameters, and the output rates varied considerably for machines with screws of the same diameter. The single screw extruders covered a wide range of screw sizes and a graph could be drawn although again the output rates varied considerably for particular screw diameters. (fig. 18

As the wide variations in output rates could have been caused by variations in die restriction, ‘output rates divided by the square of the diameter’ were plotted against ‘pipe cross sectional area’. The values used were the average cross sectional area, for each particular machine during the survey. For a particular extruder the area normally fell within fairly narrow limits owing to normal practice of changing parts within a die head whenever possible, rather than changing the complete die head. The few extruders making large diameter pipes usually had a wider range of cross sectional areas. Figures 19, 20 & 21 show that although there was a wide scatter of results, the contra-rotating twins screw and single screw extruders, had an overall trend of a gradual increase, while with the co-rotating extruders a gradual decrease existed. With the single screw extruders it can be seen that in general the size of extruder used increased with pipe size. This is a logical result as in general, when an extruder was used for a relatively small pipe

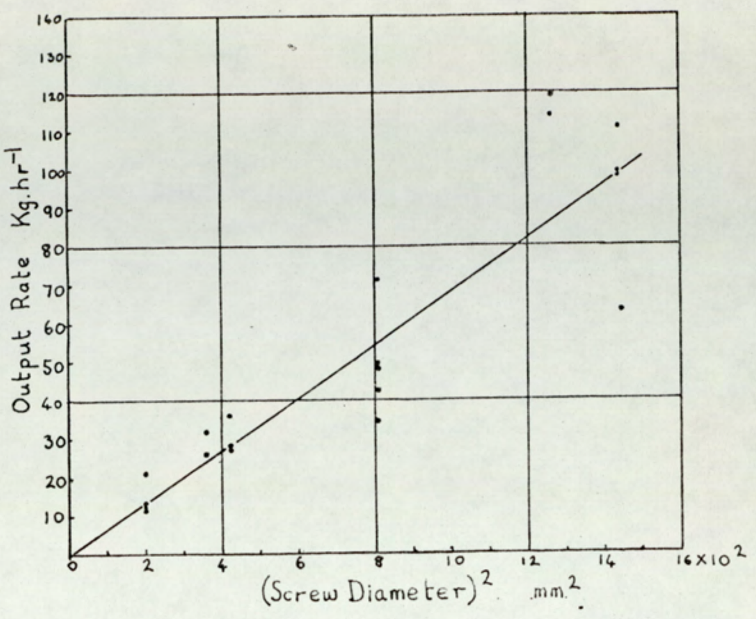


Figure 18. Output rate /screw size relationship for Single screw extruders.

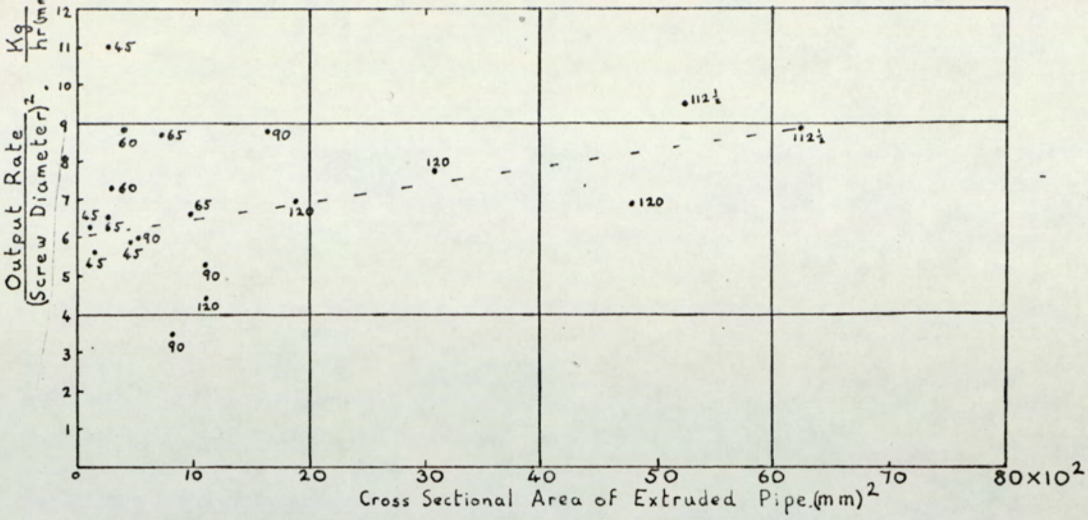


Figure 19. Dependence of output rate on pipe cross-sectional area. Single screw extruders.

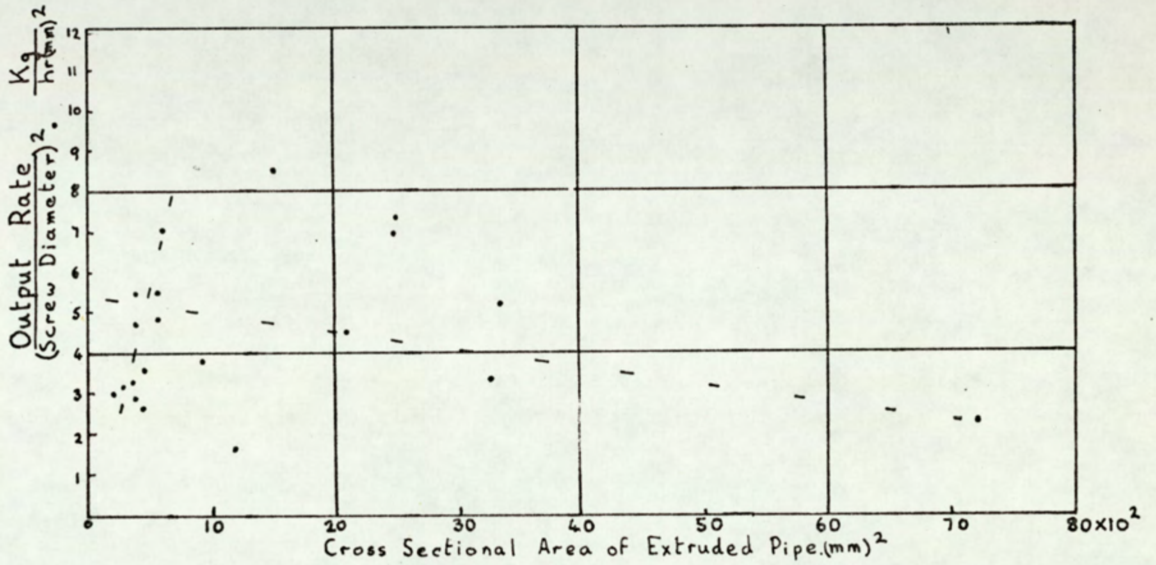


Figure 20. Dependence of output rate on pipe cross-sectional area - co-rotating twin screw extruders.

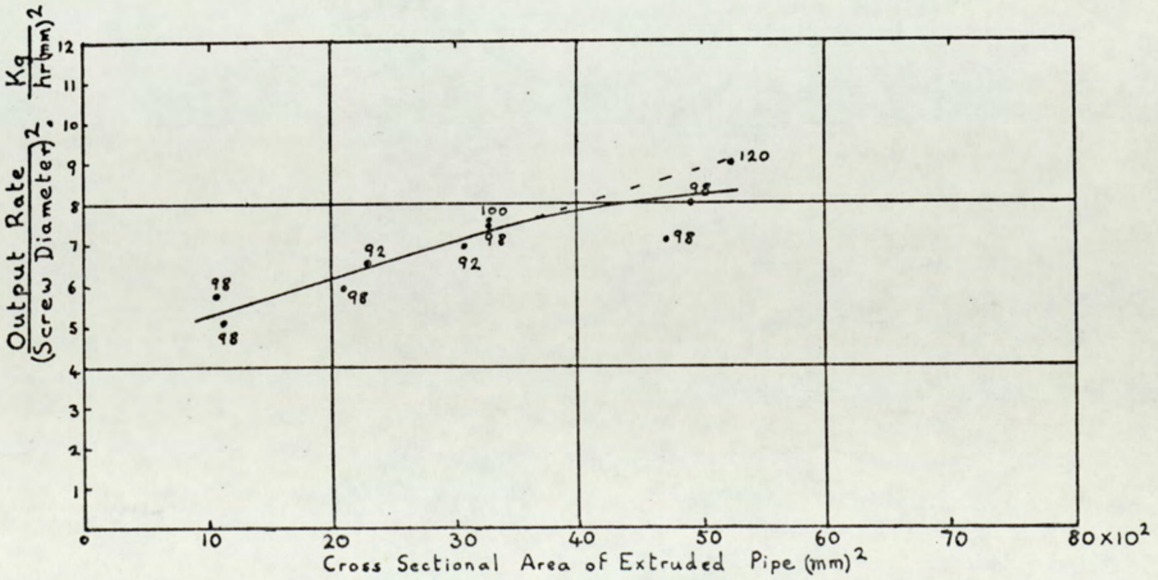


Figure 21. Dependence of output rate on pipe cross-sectional area.

size in relation to screw size, the output performance was reduced. e.g. a 120 mm extruder working with a 1.7 sq. inch area falls well below the line.

The survey showed that one of the main reasons for loss of good product was that pipe dimensions were outside specified tolerances. According to observations of the survey team, this fault, and complete breakdowns were in many cases due to poor functioning of haul-offs, saws and other ancillary equipment.

Various important conclusions can be drawn regarding production management, but from a technology standpoint, the scrap rates resulting from out-of-tolerance dimensions is the feature of most interest although waste due to thermal instability must also be considered. It is evident that close control of variables both in the production machinery and in the compounds fed into the extruder is of particular importance. As the extrusion lines could often only achieve 90% good product while running, with the figure not infrequently falling below this, then the control of tolerances appears to be sufficiently critical for significant changes in output rate to occur as a result of small variations in material properties. Control of pipe dimensions normally depends on two variables.

(i) Average wall thickness.

This depends on the relative speed of extrusion from the die and haul-off speed, and is normally controlled by adjustments to the haul-off speed control. This adjustment also allows for die swell.

(ii) Concentricity of pipe bore - (wall thickness variation around the circumference).

This depends on flow of material in the die and is normally controlled by adjustment of the position of the outer ring relative to the control mandrell. Accurate presetting of the die parts cannot

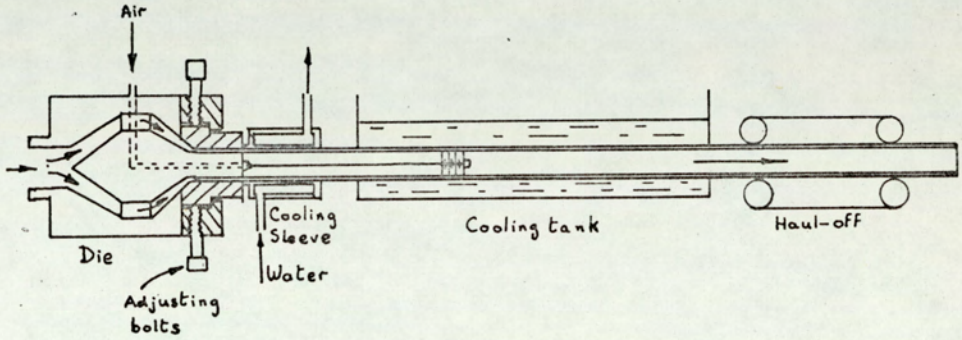


Figure 22. Control of pipe dimensions.

be relied on as variations in flow can result from slight variations in temperature as well as slight die part movements caused by the very high melt pressures which are developed (e.g. up to 10,000 psi). Slight movements will result in relatively large changes in wall thickness as the flow depends on the cube power of the die clearance.

The wall thickness of a number of commercially extruded pipes were measured at a number of points around their circumference and the results were as follows:-

TABLE 2

Nominal pipe size	Specificiation BS3505 (mm) min	max	Wall thickness (mm) min	max
$\frac{3}{4}$" Class E	1.9	2.1(1968)	2.03	2.14
3" Class B	2.9	3.3(1968)	3.0	3.28
3" Class C	4.8 3.5	5.3(1962) 3.9(1968)	4.0	4.18
6" Class D	9.1 8.8	10.0(1962) 9.7(1968)	10.55	10.87
6" Class E	12.0 10.8	13.2(1962) 11.9(1968)		

It appears that although the wall thicknesses are reasonably consistent around the circumference, none of these pipes conform to the specification for wall thickness. Even the 3" Class C pipe which had the British standard approval mark printed on it was oversize. Wall thickness will vary with output rate variations, while output rates will vary if changes in bulk density of the feed material occurs. With twin screw extruders, the "positive" pumping action of the intermeshing screws results in the output (for a given screw speed) being approximately proportional to the bulk density of the feed material. Variations in bulk density will also affect single screw extruder output as shown in Table 1.

As there was commercially a general swing to direct powder extrusion it was decided not to examine variables due to

TABLE 3 - Single Screw extruder output rates for feed materials of different bulk densities

	PVC Polymer (i)		PVC Polymer (ii)		PVC Polymer (iii)	
Particle form	Powder	Cubes	Powder	Cubes	Irregular granulate	Irregular granulate
Bulk density (g/l)	635	567	640	553	566	550
Output rate (g/min)	204	140	186	132	181	223

granule compounds but to confine further work to powder feeds. The next step was to prepare a literature review to determine what variables were already known and obtain more detailed information on the powder extrusion process. This would also avoid unnecessary repitition of work already published.

3. Extrusion direct from powder - A review

Although the main growth in powder mixing and extrusion technology was centered on plasticised PVC, direct powder extrusion probably started with rigid PVC (15). Following the introduction of high speed mixers and the relatively recent rapid growth in rigid PVC markets, direct powder extrusion of this material (adapting plasticised PVC mixing techniques) has gained considerable ground in recent years. Figures quoted for the Federal German Republic in 1967 were that of the 57000 tons of rigid PVC pipe produced, 70% was from powder (12). According to an article in 1971, (13), the proportions of rigid PVC processed from powder were 70% for pipes, 95% for injection moulding, 55% for profiles and 90% for blow moulding.

In mixing PVC, (starting from a cold powder) increasing levels of shear (friction) and temperature result in the polymer (plus additives) going through the following stages (14).

1. Slow speed mixing without heat gives a mix in which additives are distributed over the surface of the polymer particles which remain substantially unchanged; termed a "premix" or "cold blend".
2. High speed mixing, where shear and heat are applied, results in absorption of additives into the softened polymer, increased bulk density, loss of moisture and certain volatiles and improved powder flow; termed a "dryblend".

3. Further shear and heat causes further increases in bulk density followed by agglomeration and then complete gelation.

Direct powder extrusion of rigid PVC from 'premixes' on both single and twin screw extruders was described as long ago as 1949, (15) but it was not stated whether the advantages were considered to be entirely economic or whether technological advantages of such factors as thermal stability existed. 'Premixes' also feature in the latest trends in rigid extrusion development (16). A switch from "dry blend" to "premix" extrusion (made for economic reasons) has been made possible by recent extruder developments (17, 18).

Most of the literature on powder processing generally considers the advantages to be entirely economic, although admittedly often accompanied by technical difficulties such as maintaining product quality and output rate. To quote Reifenhauser (19). "The main reason for processing dryblends is the difference in price between pellets and powder, this being bigger the larger the production from unified compounds". Jones and Hawkes (14) also stress the economic reasons for dry blend extrusion as follows - " ...there is an apparent economic incentive to relying solely upon mixing in the powder form, thus reducing the number of processing stages involved between the polymer and the fabricated product". The change to direct powder extrusion was considered to have been prompted by such factors as:

(1) the need to explore all possible means of reducing the cost of the feedstock.

(2) the availability of high speed mixers of the impellar type designed to provide adequate mixing in the powder with short mixing cycles.

(3) the availability of certain types of twin screw extruder designed to handle a powder feed.

It was said that in the USA at that time (1967) most of the PVC processing (plasticised and rigid) was carried out from dryblends; single screw extruders being used, often fitted with vacuum compacting hoppers and vented barrels. European and UK developments centered around twin screw extruders.

Economic advantages listed by Lynch (20) were the elimination of expensive compounding machinery, lower conversion costs and efficiencies of materials handling.

In addition to price advantages Rabb and Ratkay (21) considered a further advantage to be that the processor could develop his own compounds without very large capital expenditure and the formulation could easily be modified to meet the requirements of the particular extruder, screw design and die.

Economic advantages were also described by Medoro (22) but he also considered that larger barrels, redesigned screws, breaker plates and dies were necessary. Malone (23) stated that high standards were difficult to maintain. Baumgartel (12) considered that an optimum degree of gelation existed and that over gelation gave rippled extrusions. Similar effects were described by Jones and Hawkes (14).

Campbell (24) gave the advantages of powder processing to be the relatively low capital investment, simplified controls and operation, degree of automation scaled to requirements, flexibility regarding materials and process, accuracy simplifying scale up and simple quality control.

The question of when to use powder extrusion is not easily found. It is evident that price advantages will only occur if quality and output rates are as high and as consistant, as for granules. In addition the capital cost of bulk handling systems which appear to be an essential part of efficient powder processing must be taken into consideration. Both Schenkel (25) and Freiman (26) point out the need for pneumatic conveying and dust hazards to health, which of course requires further capital outlay. Typical bulk handling systems have been described by Jacob (27).

Labour content of PVC extrusion is included in a paper by Ernst (6) who compared the production of rigid PVC pipes from the raw materials on ram extruders and screw extruders using both granules and powder.

Although economic advantages can be accompanied by technical problems, claims have been made for technical advantages for dry blend processing. Lynch (20) mentions reduced heat history and easier fusion. Technical advantages given by Baumgartel (12) were that products had better homogenity giving improved properties and required lower processing temperatures so that cost savings could be made from reduced levels of stabilisers and lubricants. A further advantage of powder feed was that processing behaviour between individual PVC particles could be used to accelerate or retard the gelation in the barrel and the total processing action. The product was considered more uniform as variables due to varying degree of gelation of granulate were removed. Weinert (16) considered the inferior particles flow of powders as compared with granules to be of no disadvantage as output rate was independant of this factor. The limit was dependant on heating rate of the material which was slower for granules than for powder.

Advantages of dryblends over premix given by Becker (29) were that lubricants and other melting additives were melted and homogenously dispersed, moisture removed and bulk density increased.

Cold blending could not be used where waxes had to be dispersed. Weinert (16) mentioned premixed PVC briquetted to form irregular pea shaped pieces which gave better feed properties to single screw extruders, but this process was falling out of favour due to non-uniformity.

The choice of processing method e.g. continuous direct powder extrusion, or intermediate compounding granulation, (as described in section 1) depends on both direct economic and technical factors. A summary of the general situation has been made by Reifenhauser (19) as follows:-

Mixer → extruder: very few compounds, very large outputs
Mixer → stock → extruder: range of compounds: medium outputs
Mixer → pelletiser → extruder: numerous compounds, small outputs or heavy wall sections

A few examples of when particular methods are used have been described. According to Hermann, (30) rigid pipes were generally processed directly from PVC dryblends or agglomerates, which according to Dahlmann (31) was for economic reasons. In cable coating the granulation step was said to be almost invariably used (30) being attributed in another paper to the technical problems of feeding and plasticisation as a result of high speeds and short residence times. However dryblends could be satisfactory for heavy wall sections. Granulation was also considered (by Hermann) to be generally used for preparation of high impact PVC where perfect homogenity of polymeric additives in the PVC was necessary.

For profile extrusion, the economics of changing from granules to powder were stated by Dahlmann (31) to be often unfavourable, and for single screw extruders (quite frequently used for this application) the change is only occasionally made. There is the possibility of reduced output and higher labour costs as well as unsuitability of dies due to different swelling behaviour.

Dies for forming thin wall electrical insulation tube (conduit) produced high back pressures which allowed single screw extruders to be used.

Hermanns conclusions (30) concerning the question as to which of the process methods offered the greatest advantage were "...that the answer must be found individually for each case and is particularly influenced by the formulation employed, the application of the semi-finished or finished part and the technology of the equipment used in processing."

3.1 "Slow speed" mixers.

Engels (32) states that the production of dryblends involves heat input which can be by conduction, convection, friction, mixing friction and preheating. Various trough mixers using indirect heating involve conduction and convection processes. The production of plasticised PVC dryblends in various types of mixers has been described (33) giving some indication of the advantages and disadvantages of each type. The mixers described include ribbon blenders, rubber type mixers, conical mixers and dough mixers.

The earliest published literature is concerned with the production of plasticised PVC dryblends in ribbon blenders mainly for cable production (34, 35, 36). The general procedure has been described by Freiman (26) in which a two stage double spiral ribbon blender was used. The method, using a blade rotation speed of 60 to 80 rpm was to heat and mix the polymer and stabilisers for ten minutes, and then the plasticisers, which had been pre-heated to 80 to 90°C were injected through small holes in a central distributor. Although cold mixing was possible, pre-heating increased plasticiser absorption and decreased the cycle. The mixing was continued until a well dried powder was obtained. The mixing cycle could be followed by recording motor current. The powder reached a temperature of 110 to 130°C in a cycle time of 30 to 34 minutes. The drying time depended on the plasticiser absorption by the polymer. A polymer with very good absorption could give a very dry powder with some plasticiser-free particles which would subsequently produce "fish eyes", so a compromise had to be made to give reasonable cycle times.

The hot material was then transferred to a second mixer which was water cooled, so that the material, which would otherwise have stuck together and decomposed remained in a free flowing condition.

This procedure appears to be typical for dryblend preparation in ribbon blenders although there are some variations. The method used by Flathers (37) was to add the plasticiser at room temperature, apply steam heating after 5 minutes mixing, lubricants being added after the mix had become free flowing, and finally discharged to a cooler at 100 to 105°C. The procedure described by Medoro (22) was to heat the polymer in a ribbon blender to 80-90°C with the jacket at 115-120°C, liquids being sprayed in and then dry ingredients such as stabilisers and fillers were added. Plasticiser preheating has been advised for mixes with high plasticiser concentrations (e.g. 80 parts per 100 PVC), while viscous plasticisers need higher mixing temperatures (25).

The production methods for plasticised PVC are of interest as they have been generally applied to the relatively recent dryblend mixing processes for rigid PVC compounds. In this case the considerations concerning liquid additives such as liquid stabilisers and sticky process aids can often be regarded as analagous to problems of plasticisers in plasticised PVC.

Engels (32) gives batch mixing times from 15 to 240 minutes depending on the nature of the material. When a certain amount of unavoidable frictional heating occurred at narrow lands between mixer blade and trough it was regarded as undesirable, being non-uniform which could cause "fish-eyes". Slow speed dryblend mixers are used for mixing very large batches of material and use ribbon blenders for suspension polymers and double bladed sigma blade mixers for emulsion polymers. Medium speed dry blend mixers are a similar type of machine run part filled and at a faster speed to throw a small proportion of the mix into free areas. Mixing time is reduced by 25% but this is offset by the reduced load. Ribbon blenders can be used for continuous mixing, but problems occur in controlling dwell time.

Interest in ribbon blenders is being renewed by the development of cold blend extruders for certain applications as the capacity of this type of mixer can be very large.

3.2 High Speed mixers

The ribbon blender is limited by long cycle times resulting from its reliance on transfer of heat from the jacket rather than heat internally generated by friction.

The principle of using friction, as used in the "Henschel" high speed mixer was described by Piltz (38) in 1954, where mixing and stirring of liquids was used as an analogy. In liquid mixing processes, slow speed stirrers with peripheral speeds of 3 m/sec could be replaced by reduced diameter stirrers with peripheral speeds of 20m/sec. (Typical rotor speed given as 1800 rpm). For mixing powders, the high speed mixer uses the same principle whereby high peripheral speeds are used and high velocity gradients created. The material can be treated as a liquid as parts of the rotor aerate the material and this aerated or fluidised powder flows downwards into the rotor, becomes subject to shearing forces and returns upwards giving a vortex similar to that of a stirred liquid. (29, 38). Air for fluidisation is obtained by drawing it down at the centre of the rotor (24, 38). Due to this aeration, peripheral speeds of up to 40-60 m/sec can be used (24, 38, 39) and power requirements are not excessive (38).

Most high speed mixers are basically cylindrical in shape with the height equal to the diameter and with a rotor as near the base as possible (39). Construction details have been described by Becker (29).

The frictional heat is developed by the mixing action. The high rotor speed applies impact and friction forces tangentially such that these and radial centrifugal forces throw the compound against the wall where it is deflected upwards. Particles are rubbed together and in a fluidised stale form a vortex (29). The rapid circulation of material creates differential speeds between particles and mixing blades (rotor) vessel wall and other particles which develop the frictional heat (32).

The frictional heating process was claimed (38) to be quicker and more economic than other methods as polymers have low thermal conductivity and there is poor heat transfer between the polymer and the walls of the mixer. Also, being a direct heat source it can be well controlled, and the required temperature can be attained in the shortest possible time as a result of the frictional heat developed (32).

The high shear gradients and shear forces are such that in addition to dispersion processes, comminution and distribution of stabiliser and pigment agglomerates can occur (30). Claims are also made that it is possible to mix PVC with up to 50% plasticiser 0. 02% pigment and 0. 01% blueing agent (40). The heating received during mixing promoted drying which could be further improved by using a vacuum. As many products bridge in high speed mixers, doctor blades and scrapers have been developed to avoid the condition where a ring of compound clings to the wall by centrifugal force and fails to fall into the central rotor. A doctor blade rotating at slow speeds was considered to be superior to a fixed scraper (32).

Procedures for dry blend mixing of both rigid and plasticised compounds appear to be similar to those for ribbon blending. High speed mixer discharge temperatures appear to be normally in the range 80 to 150°C (38, 40, 41) taking between 5-20 minutes to reach this temperature. The hot mixer is discharged into a slow speed cooler (ribbon blenders can be used). The cooling, which is to 30-60°C, should match the high speed mixing time. The construction of the cooler is usually similar to the high speed mixer, except that it is 2-3 times larger to achieve adequate cooling. It has simple blade design, low power motor, water jacket cooling, and in some cases, water cooled rings or blades (29). Stirrer speeds of about 200 rpm are used (24).

Development have been aimed at shortening cooling time to match high speed mixing time. Systems used are increased diameter in relation to height to increase surface area, cooling rings and cooled stirrers (39). Filtered dry air reduces condensation on the mixer wall (24) but oxidation of the PVC may occur (34). Discharge temperatures below 60°C disproportionately extend the cooling time (39).

3.3 High speed mixing of unplasticised PVC

Most of the literature on high speed mixing is concerned with plasticised PVC presumably because this material offers the biggest problem in obtaining a free flowing powder, particularly when high plasticiser levels are used.

With the absence of plasticisers, the problems of maintaining the mix in a free flowing condition and absorption of plasticiser to produce a free flowing powder often no longer exist. A typical cycle involves reaching 120°C in 8-9 minutes so that moisture can be evaporated and lubricants and stabilisers melted for more uniform distribution, theoretically covering each particle. The mix is then cooled to 30-40°C (31). In order to reduce the mixing time, lubricants can be omitted till near the end of the high speed mixing stage to increase friction and so speed up the heating rate (21).

However, due to the critical nature of the extrusion of unplasticised PVC dry blends, the condition of the dry blend and consequently the mixing condition can become quite critical. Interaction between the process of dryblend preparation and extrusion process is such that any change in the stage to which dry blending is taken will have a direct effect on extrusion performance (14). Basic controls are temperature, time and power input, but temperature is normally used for process control (32). Becker (29) considered that preliminary tests must be used to determine the discharge temperature required for optimum increase in bulk density, which varied with polymer specification.

The effects of the rise in temperature during mixing have been described by Guimon (42). Up to 80-90°C additives are dispersed. Between 90 and 120°C absorption of additives starts and at 120°C small particles gel this becoming progressive with increasing temperature until at 160-180°C decomposition starts. At the elevated temperatures, PVC absorbs stabilisers and lubricants in a similar manner to plasticisers and thermal stability is superior to cold blend mixes.

There appears to be general agreement between the results of Guimon (42), Jones and Hawkes (14), and Morohashi (43) on the effects on the polymer of high speed mixing to elevated temperatures. Photomicrographs show that particles were opaque up to 100°C. At 120°C (42) to 150°C (43) smaller particles "gelled" and became transparent, while larger particles became progressively transparent with further rises in temperature until at 160°C most were transparent (14) and at 170°C practically all were transparent (43). At temperatures up to 120°C, opaque particles were relatively few, but some densification occurred, attributed to lubricant dispersion (14).

Graphs of power input and temperature against time show an initial fall in motor current followed by a rise passing through a peak. Morohashi's results showed that as the mix temperature rose, the motor current fell at 50°C passing through a minimum at 80-110°C, then rising rapidly, while Guimons results showed a power decrease occurring up to 90°C which was attributed to lubricant dispersion effects. Differences would therefore occur with different formulations. According to Morohashi the decrease corresponded to swelling of the PVC. The rise in power between 90 and 120°C corresponded to the PVC being above its softening point so that lubricants could be absorbed (42). The fall in power beyond the peaks according to both Guimon and Jones and Hawkes was due to completion of wax absorption which would also reduce tackiness but could be due to formation of more regular particles (14).

Increases in bulk density with discharge temperature and the consequent improvement in extruder output rate are shown by both Guimon (42) and Jones and Hawkes (14). The effect of increasing discharge temperature was to increase bulk density, output rate and falling weight impact strength. Over a range of mixer discharge temperatures from 110 to 160°C, bulk density increased from 699 to 870 g/l, output rate for a twin screw extruder increased from 178 to 208 lbs/hr and impact strength from 36 to 103 ft/lbs (140°C). The exception of reduced impact strength for 160°C material was attributed to the sieving out of agglomerated additives. Guimons graph of bulk density against mixer temperature shows rises from 0.65 at 100°C to 0.080 at 180°C. The corresponding graph of extruder output rate shows rises from 50 to 65 kg/hr. In a "non-toxic" formulation, bulk density increased from 0.67 to 0.71 to a mix temperature of 140°C, which increased extrudate impact strength from 45 to 51 kg/hr. Impact strength reached an optimum at 90 and 120°C.

Morohashi (43) associated bulk density increase with particle size reduction. The particle size distribution showed an increase in the number of small particles (170-180μ) mainly at the expense of the 100μ size which constituted most of the mix at the lower temperatures. Guimon (42) showed that small particles tended to agglomerate either together or with larger particles to change size distribution. The effect was that fine particles, which otherwise cause problems in single screw extruders are removed, and output increased as less energy was required to melt gelled particles. However Jones and Hawkes (14) considered that power requirements are increased.

3.4 Polymers

The application of DIN test methods to characterise PVC polymers has been described by Frerichmann (44). Comparisons of emulsion, suspension and mass (block) polymers are made, although for many properties little distinction is made between suspension and mass polymers.

A basic difference for dry blending properties is that the particles type and size of the suspension polymer are determined by the polymerisation conditions, while with the emulsion polymer they depend more on the processing of the latex. Suspension polymers can be smooth and glassy, or irregular and fissured. A particularly important point is the relationship between the bulk density and the K value. The K value is a constant derived from solution viscosity measurements and can be taken as a measure of the polymers molecular weight. As the K value is increased, the bulk density decreases, or if maintained, results in particles becoming more glossy. The consequence of this is that for higher bulk density (for higher output rate , better absorption of plasticisers and other liquid additives and therefore faster gelling rates) a low K value is preferred. However, a higher K value gives improvements in tensile strength, and fatigue strength, higher softening point, better thermal stability and lower gas permeability. (Presumably the K values normally used are a satisfactory compromise). The residual emulsifiers in emulsion polymers were said to make processing easier but have an adverse effect on the final product as a result of water absorption. Emulsion polymers normally contained small particles and as a result had poor particulate flow properties and also had this effect when mixed with suspension polymers, countering the increases imparted to bulk density.

With emulsion polymers, the initial finely divided particles from the polymerisation stage are agglomerated by either electrolytic precipitation, spray drying or roll drying. Only roll dried and certain spray dried types had good flowability. In general fine powders caused poor flow properties. A narrow size distribution range was required as a higher proportion of larger or smaller particles reduced flowability. Fine particles could occupy the space between larger particles to increase bulk density but flowability was reduced.

According to Dahlmann (31), for uniform free flowing blends, particles less than 60μ should not exceed a level of 15%. A big increase in the proportion of fine particles can result from badly worn mixing blades. For pressure pipes, suspension polymers were normally used. If more than 20 phr emulsion polymer is used then the DIN water absorption tests are exceeded. With the older types of twin screw extruders the polymer density should not exceed 560-580 g/l which gives a mix density of 620-640 g/l. With newer machines, 640g/1 polymer can be used. Jones and Hawkes (14) considered that the"mild working" of material by a low compression extruder required irregular particles to generate inter-particle friction to ensure gelation. Higher compression ratio extruders with kneading zones, etc. can take full advantage of higher bulk density polymers.

However difficulties can occur as explained by Dahlmann (31) in that high bulk density polymers have harder glassy particles that are difficult to melt and that usually the same polymer has to be supplied to the whole production covering a range of ages and types of extruders. This same consideration applies to formulations.

For other applications Dahlmann's recommendations are as follows:-

For profile extrusion for indoor use, an emulsion polymer with a K value of 70 is preferred to a suspension polymer as the better surface appearance required by applications such as furniture can be obtained. The emulsion polymer should be free flowing with a 60-70 K value.

For electrical insulation tube, rapid cooling of thin walls made high extrusion speeds possible, implying short dwell time and high friction so that mechanical strength was attained with 60 K value polymer.

For cables polymer to plasticiser ratios of 60/40 - 75/25 could be run.

Morohashi (45) examined the effect on dry blend bulk density and mixing time due to polymer particle size and bulk density. Five suspension polymers covered high and low bulk density, fine and large particles and a standard grade. Three bulk polymers covered fine, large and standard particle sizes. The effects of mixing to 150°C on bulk density were as follows: For the high bulk density polymer, the bulk density was little changed, even though motor current was high. The bulk density of the low bulk density polymer was significantly increased, while the large particles (lowest bulk density) also had a significant increase in bulk density but with no significant increase in motor power. With small particles, mixing action was poor and bulk density was not increased. Mixing times to reach 145°C were 9 minutes for high bulk density polymer and large particle polymer, 10 for the standard and 12 for low bulk density and small particle size polymers.

Gelation tests were carried out on a two roll mill using dry blends prepared from narrow sieve fractions giving a range of bulk densities from 0.748 to 0.69 and also using pellets (43). The results showed gelation time to be in the order of bulk densities, 0.748 gelling first and pellets last.

3.5 Stabilisers

Information on PVC stabilisers is usually of a general nature equally applicable to any form of PVC processing. Heilmayr (41) lists the usual requirements for the ideal PVC stabiliser. These are, efficient stabilisation, good colour, low volatility, odourless, low toxicity and of course, low cost. Mention is made that barium-cadmium mixed stearates have insufficient stability and that a small addition of lead stearate is necessary.

Lang (46) considered that stabilisers have little influence on dryblend properties, except when liquid stabilisers are used in unplasticised PVC. In this case the required powder flow can usually be obtained by increasing the mix temperature.

According to Dahlmann, (31) stabilisers present difficulties only in exceptional cases and selection was more concerned with such factors as meeting toxicity regulations.

3.6 Lubricants

It appears from a number of papers, that as described by Lang (46), lubricant selection depends on processing conditions and the influence of other additives on compatability. It is normally necessary to use a combination of several lubricants.

Lubricants were classified chemically into three basic types; pure hydrocarbons, substituted hydrocarbons and fatty acid derivatives. Classification according to effect divided them into two basic groups of external and internal. External lubricants imparted release properties following formation at the plastics surface, while internal lubricants, as a result of their good compatability, reduced the melt viscosity. It appears from this and other papers (28, 51) that the term "external" is applied to lubricants of low compatability with the PVC, while intermediate types also exist.

A table is given of these two basic types. Their influence was said to depend on compatability, melting behaviour and amount used. Jones and Hawkes (14) considered that incompatible lubricants with low melting points had the greatest effect. Twin screw extruder results showed that concentration changes of stearic acid from 0 to 0.33 phr produced extrudate changes from overworked to ungelled. Wax E, which was considered a partially compatible lubricant showed a less pronounced effect.

Accumulation of incompatible, sticky or liquid lubricant on the surface of the PVC particles will impair flow properties, while the melting point should be such that the lubricant melts (46). Consequently it will then be well distributed but solidified when discharged from the cooler (about 40°C). A combination of compatible and solid external lubricants in the required melting range should be used.

This is further explained by Jones and Hawkes (14). A result of absorption and adherence of lubricant to PVC polymer surface during high speed mixing is the inhibition of gelation due to interference with inter-particular friction. The effect of lubricant on gelation behaviour was considered more important than effects on melt flow behaviour.

Heilmeyer (41) classifies stearates of fatty acids and fatty acid esters as "internal" and low molecular weight polyethylene lynisolen, and stearic acid as external.

Twin screw extruder results of Morohashi (47) included effects of different levels of two lubricants on physical properties. Lead stearate over the range 1.8 to 2.3 phr showed a maximum tensile strength at 2.0, while elongation fell. Stearic acid over the range 0.1 to 0.5 showed slight falls in both properties up to 0.3 phr, falling more steeply above this level.

According to Jones and Hawkes, lubricants having the most significant effect on gelation were those active between 90 and 100°C as PVC particles soften in this range. A method demonstrated of giving precise control of gelation rate was to use mixtures of normal and dibasic lead stearate (total 0.75 phr) the former type being more efficient than the latter. Increasing the proportion of dibasic lead stearate increased motor current and decreased impact strength once an optimum at 0.65 normal lead stearate 0.10 dibasic lead stearate was exceeded. At 0.75 phr normal lead stearate (zero dibasic lead stearate) impact strength was only 20% of the optimum. These two materials formed the main lubricant, but the additional incorporation of stearic acid or wax E was considered necessary to provide greater gelation control. Increases in level of either of these lubricants would move the gelation point along the screws towards the die, reducing both motor current and "working" of material. Wax E was preferred to stearic acid as its action was "milder".

It was suggested that lubricants were effective up to a temperature 50^{o} above their melting point. For the temperatures of around $180^{o}C$ reached in the die head, Calcium Stearate was recommended. A similar type of formulation is given by Baumgartel (12) in which stearic acid is used instead of Wax E. His graphs of changes in output rate with increasing lubricant level showed different behaviours for different materials. The relatively incompatible materials, paraffin wax and stearic acid, showed a rapid initial drop in output from (0-1 phr lubricant) but then fell more slowly. Glycerol monoricinoleate and a fatty alcohol (relatively compatible with PVC) showed an initial increase (from 0-1 phr) and then a relatively slow fall with further increases in lubricant level. Lead stearate and wax E showed quite sharp increases in output rate with a peak at 0.5 phr but then fell quite quickly; wax E in particular giving the lowest output of all the lubricants when at a high concentration. Power consumption curves followed the same trends. These results, which were obtained on a 30 mm single screw extruder were for lead stabilised formulations. Behaviour of lubricants was said to vary widely with stabiliser, so the desired lubricating action with certain stabilising systems were not obtained in others.

Advantages claimed for powder extrusion over granule extrusion were that lower lubricant concentrations were required and that gelation rate could be controlled by the use of such additives.

In selecting a lubricant other factors had to be considered such as the effect on thermal stability, electrical properties, adhesion, etc. The effect of lubricant on the transparency of extruded products can also be important (41).

3.7 Fillers and polymeric additives

Fillers and polymeric additives are used at levels up to 10 phr to improve processing and/or impact strength.

The fillers normally used are stearate coated precipitated calcium carbonate or finely ground chalk. Their use has been described (48, 49) but the reasons for their improvement in impact strength has not been explained. Lang (46) considered the effects of fillers on dry blend properties. Treated chalk gives improved flow, increased output rate and cleaner surfaces. Other fillers were considered mainly in consideration of product properties.

With plasticised mixes fine fillers had deleterious effects on flow, while porous fillers, although having an undesirable plasticiser absorption effect, did provide drier and therefore better flow mixes. Baumgartel (12) considered that poor flowability resulting from fine particle additives such as fillers and impact modifiers can be overcome by either changing mixing conditions, reducing additive level, increasing particle size, or using bonding agents to reduce dust level. Fillers are not allowed by DIN (German standards) in pressure pipes, but are allowed in waste and drain pipes. Levels limited to 3 phr are allowed in sewer pipes to make processing easier. Jones and Hawkes (14) showed that increasing the filler concentration caused a fall in output and increased melt viscosity in twin screw extrusion. 10% filler level gave some decrease in gelation time. Falling weight impact tests showed an optimum at 7.5 phr while notched bar tests showed a continuing increase up to 10 phr. The difference was attributed to flaws in the extrudate caused by the high filler loading, needing reformulation to increase mixing of the filler. According to Engels (32), although fillers increased power requirements, higher levels could be used when using the dry blend process. Morohashi's twin screw extrusion results (47) showed that with one chalk filler, tensile and elongation properties were about the same at 10 phr as at zero phr, passing through a minimum at 5.0 phr, while with another chalk filler the elongation and tensile strength fell with increased filler content. It appears from the text that the former one was uncoated and the latter one stearate coated.

Polymeric additives include the copolymers methacrylate butadiene styrene, acrylonitrile butadiene styrene, ethylene vinyl acetate (66). Chlorinated polyethylene and various rubbers are also used (67). They appear to be dispersed as a separate phase thus forming a blend with the PVC polymer

Certain chlorinated polyethylene and ethylene vinyl acetate impact modifiers require the intensive gelation attainable only in a granulate stage (12) for adequate dispersion.

Acrylic modifiers at a level of addition as low as 3 phr can impart a smooth glossy surface, but large particle types should not be used for dry blends (41). Brabender plastograph results by Jones and Hawkes (14) indicate a considerable shortening in gelation time with as little as 2% addition of process aid and a far greater change than that for the addition of 10% fillers.

3.8 Pigments

The selection of pigments with regard to processing from dry blends has been described in detail by Herrmann (30). During high speed mixing, inorganic pigments and milled pigments are distributed while agglomerates are partially broken down during subsequent processing. Difficulties can occur where high shading accuracy is required, although masterbatches can be used if particle size is correct. In general, solids should be fed first and well mixed before liquid addition to prevent formation of pigment lumps.

3.9 Extruders

Ram, single, and twin screw extruders can be used for rigid PVC extrusion. The ram type appears to be the earliest extruder used for rigid PVC but requires the PVC to be milled and rolled up into "dollies" for charging the extruder (6, 15). This machine was never widely used, being superceeded by screw extruders. Both single and twin screw types were used for granule extrusion (see Section 1). With the change to powder extrusion, twin screw extruders have predominated in Europe, while in the USA single screw extruders are favoured. (14).

3.10 Single Screw extruders

In an early description (1949) of powder extrusion (15) confidence was expressed that, with experience, feeding of unplasticised PVC as a premixed powder to a single screw extruder would be common practice. It was considered that both a conveyor allowing premixed polymer to be heated to about 100-120^{o}C prior to the feed, and an extruder screw with a smear head for efficient mixing, were both necessary.

The advantages of single screw extruders are that they have a heavy duty construction, high thrust bearings give high output rates with minimum "down time" while capital and maintenance costs are lower than for twin screw types. They are considered more versatile, while the absence of feed metering mechanims (frequently fitted to twin screw extruders) is less to go wrong. (50).

The back pressure produced by the die needs to be at a high level for shear stresses in the barrel to be high enough for conversion of powder to a melt. According to Dahlmann (31) the lower back pressure produced by larger pipe dies are insufficient to achieve conversion of powder to a melt. Provision of restrictions in the die head to overcome this deficiency reduced the output rate. The applications of single screw extruders were said to be limited to pipes up to 50 mm diameter, blow moulding, moderately heavy profiles and light profiles. For profiles, 45 and 60 mm extruders could be run at 20 and 30 kg/hr respectively. The necessary back pressures for profile extrusion could be obtained by using 'breaker plates' with 2-3 mm holes.

The limitations of large dies was also given by Marahashi (43). He also considered that the varying level of powder in the feed hopper, resulting from the absence of a meter feeder, varied the bulk density of the powder at the hopper base and hence varied the output rate. An example was given of a 125 mm single screw extruder producing satisfactory corrugated sheet but using dryblends discharged at 175^{O}C.

In considering single screw extruder design, Lounsbury (50) gives a screw L/D limit of 24:1 as if a 32:1 screw appears necessary it is simpler to use a 24:1 extruder of the next largest size. However a size limitation for rigid PVC was considered to occur at 6", (150 mm) which imparted too much work, resulting in thermal degradation. $3\frac{1}{2}$" (90 mm) and $4\frac{1}{2}$" (115 mm) were considered suitable. A depth ratio of 3:1 was suitable for dryblends. An output rate limit was imposed by entrapped air if screw compactors and vacuum (700 mm Hg) were not used while cold blended materials must be degassed to remove volatiles (52). Vacuum can be applied either at the hopper, or at a vent along the barrel providing a two stage screw is used. Weinert (16) considered barrel venting to be easier than hopper vacuum as powder can block the vacuum system. However barrel venting is more costly (52). In one example (52) output rate was increased from 44 to 75 Kg/hr, by hopper vacuum, but the preferred method was forced feed combined with a vented barrel, which with 90 mm and 115 mm extruders had produced rigid PVC pipes at 80-90 and 135-180 Kg/hr respectively.

Lounsbury considered that a screw depth ratio of 3.0:1 was satisfactory for dry blends but even higher ratios have been recommended (19). Marahashi (43) considered 3.5:1 excessive as it reduced output rate. However this applied for the exceptionally high dry blend bulk densities (0.75) recommended by Marahashi. If mixing conditions were such that dry blend bulk density was lower (e.g. 0.65), then performance of the single screw on powder was inferior to that on granules. With the high bulk density dryblends, the general requirements for single screw extrusion concerning compression ratio and metal soap lubricants were considered invalid as were claims for merits of granules as against powder.

Menges and Klenk (9, 10) carried out experiments on the melting mechanism of rigid PVC dryblends in a single screw extruder and compared their findings with those of Street (53) and Maddock (54). Although rigid PVC was not included in Streets experiments and only briefly mentioned by Maddock, it is necessary to consider their findings in some detail as they illustrate how a single screw extruder normally functions as a melting and conveying device while a different mechanism was shown by Menges and Klenk to occur when PVC dry blends are used.

In Streets experiments (53) an extruder was fed with a mixture of coloured granules and the material in the screw channel was examined following its removal from the barrel. Polymers used were polyethylene and plasticised PVC. The flow patterns of the coloured material showed that the fusion action was as follows. As the granules are conveyed into the mixing zone the first material to melt does so against the barrel wall and the following flight scrapes it off, so forming a rolling mass in this region. As the material travels down the screw channel there is a continuous addition to this "melt pool" by that being scraped by the flight from the barrel wall, as well as that erroded from the solid granules by the circulating melt pool. As the molten material increases in proportion to the total cross-channel area the area of granules exposed to the barrel surface decreases and this contribution is decreased. The rolling melt erodes little from the granules at first, but a constant area is exposed once it reaches the full channel depth.

Another set of experiments showed differences in the conveying rate of granules compared with molten material. The procedure was to add granules in increments just sufficient to fill the screw channel for one turn, each succeeding increment being a change in colour. Examination of the melt in the screw channel showed that the granules were conveyed faster than the molten material. This meant that the voids that would otherwise be left by the granules that had been melted down were filled by granules which were travelling faster than the melt. In comparing granules with powder, air resistance was low between 1/8th inch granules and good evacuation could take place, but with fine powders there was a considerable pressure drop which impeded flow of the powder. The overall effect was that the powder which fused first took far longer to travel the length of the screw than that fusing last.

Maddock (54) carried out practical investigations using a screw removal procedure similar to that of Street. Points by Maddock, in addition to those already by Streets papers, which are concerned with mixing and lubrication are as follows. In some cases, some of the melted material travelled forwards across the width of the channel between the unmelted material and a screw root, to be returned to the barrel surface at the trailing edge of the forward flight. It was then transferred back to the rear of the channel via the barrel surface, where it presumably as set down as a skin and scraped off by the following flight. Overall flow patterns will be in a helical path, which contributes to mixing efficiency. The effect of lubricants was to reduce adhesion of material to barrel and screw surfaces which delayed "plastifying", while in extreme cases circulating flow in the rear portion of the channel did not occur and neither did any mixing action take place.

In Menges Klenk's experiments (9, 10) a number of commercially produced unplasticised PVC dry blends were used, while the majority of the experiments used Solvic VP1710/1 which was based on a polymer with a K value of 64 and a bulk density of 0.55; average particle size 0.110 mm. The experiments showed a different fusion mechanism to occur to those of Street and Maddock described above. This difference was attributed to unplasticised PVC being non-wetting, in contrast to wetting thermoplastics such as polyethylene. PVC powder in contact with heated surfaces became sintered together and flowed down channel without adhering. Pressure due to screw conveying and reducing channel depth forced the material into the clearance between flight land and barrel wall, where it was intensively sheared into a molten state. This material collected at the forward side of the channel and at the end of the metering zone occupied 10-80% of the channel volume depending on the extrusion conditions. PVC powder remote from the barrel surface was sintered together and was discharged by the screw in an undermixed inhomogenous state. Air was trapped in the central area. Summarising their experiments, Menges and Klenk considered that the "plasticisation" method for single screw extruders was different for adhering materials than for non-adhering materials. In the latter case, material subjected to the clearance between flight land and barrel surface was exposed to a high shearing force which produced a homogenous material while the rest was heated by conduction and convection only and not homogenised at all. Temperature differences between these areas were up to 15°C. From these results, it was concluded that economic extrusion direct from powder on a single screw extruder was not possible.

The fusion model of Menges and Klenk was considered by Wusten (52) to explain why venting was unnecessary with granules but necessary with powder. A screw which operated satisfactorily on granules could not be expected to produce the same output rate and extrudate quality from powder. As flight depth ratios vary considerably between extruders, the processor usually matched the lubricants to the screw used, but the flight depth in the metering zone must not be too shallow.

In a direct comparison between single and twin screw extruders (68), the same die and same dryblend were run on both types of machine. The results for the 140 mm dia. thick wall pipe showed the creep strength of that made on the twin to be the better of the two.

Twin screw extruders

As this review is concerned with single screw extruders it is not proposed to treat the subject of twin screw extruders in detail but only to include data relevant to processing differences. Although twin screw extruders are used in preference to single screw machines for processing rigid PVC dryblends, there is less published information than on single screw types.

The output rate of a twin screw machine is not as dependent on temperature as a single screw machine so that the dimensions of the extrudate are more constant. For this reason, they are favoured for thick wall large diameter pressure pipe. (41, 50). According to Heilmeyer, (41) twin screw extruders give good mixing of the material at low back pressures which is important for processing dryblend and avoids the major problems of pressure build-up necessary in single screw machines. The large shear forces between the intermeshing screws break down gelled particles, giving superior crystal clear articles. Co-rotating screw generate more shear forces than contra-rotating screws so that running conditions and raw materials are rather more critical with the former type. A disadvantage of twin screw machines is that the screws leave marks on the extrudate, but this can be overcome by using an insert with a small cross section. An argument used against twin screw machines is that bearing sizes are limited and are therefore easily overloaded, but developments in bearing design were claimed to have largely solved this problem. (41).

In addition to a choice of counter and co-rotating screws, flights can have different and varying configurations. Compression can be obtained by decreasing the pitch, increasing the core diameter or by increasing the width of the flight (7). Twin screw extrusion has been described generally by Schultz (55), Domininghaus (56), Marhenkel (57) and Zielenhowski (7). General requirements concerning degassing, metering of the feed, screw temperature control etc., have been described by Weinert (16). Powder extrusion on a co-rotating twin screw machine has been described by Kistler (58) while Doboczky (59) has produced a detailed article on theoretical aspects of mixing behaviour of counter and co-rotating screw machines and compared them with a single screw extruder.

4. Factors involved in high speed mixing

The first part the process examined was the preparation of extruder feed material by high speed mixing. From examination of the literature on the flow and feeding of powders it was evident that variability in this stage could be very significant. It was therefore desirable to know what effect the discharge temperature from high speed mixer to cooler had on the physical properties of the PVC powder and hence its subsequent extrusion behaviour. With most suspension polymers, the heat developed in a high speed mixer is mainly due to friction and not due to heat transfer from the heating jacket of the mixer as shown in Fig. 23.

Jacket heating is usually necessary with production size mixers to ensure the mixer is at a consistent starting temperature and to reduce heat losses from the powder.

Lubricants will influence frictional heating and will therefore be important. The number of materials available as lubricants for PVC extrusion is very large and vary considerably in type; unfortunately very little useful information is normally supplied on their extrusion behaviour. In this study a few representative lubricants have been taken to show how they can effect material behaviour and the tests applied are described. By using a combination of a relatively compatible lubricant with one of relatively low compatibility in various ratios, a range of lubrication efficiencies and processing behaviours can be obtained (14). In this work Glycerol mono-stearate GMS and stearic acid were used as compatible and incompatible lubricants respectively. Both materials were used as commercial grades.

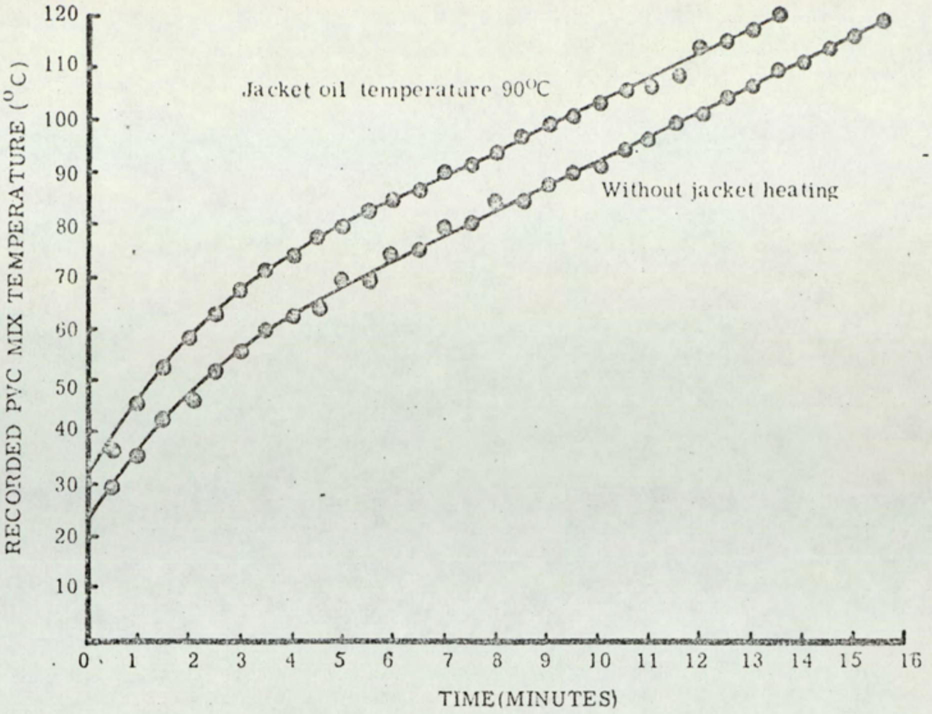


Figure 23 - Effect of jacket heating on rate of temperature rise during high speed mixing

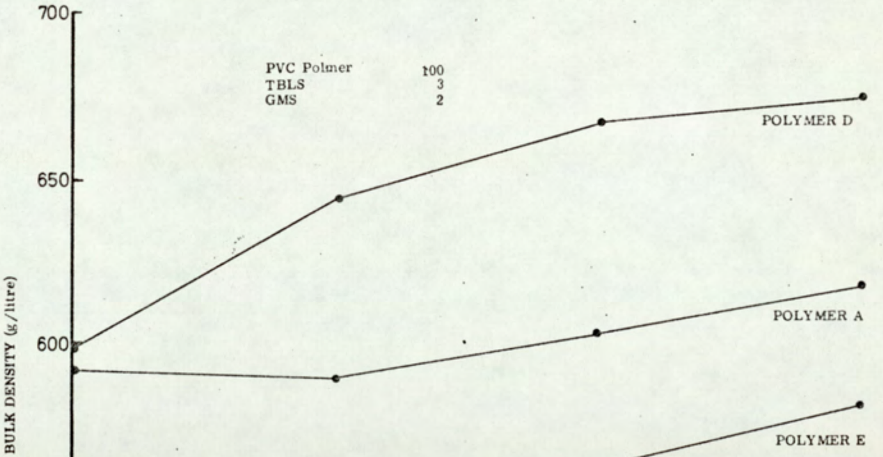


Figure 24 - Change in bulk density with mixer discharge temperature - Comparison of three polymers

4.1 Effect of mixer discharge temperature

According to published literature the possible advantages of discharging at as high a temperature as possible are as follows:-

(1) Increased absorption of stabiliser giving improved heat stability.
(2) Increased bulk density of the powder.
(3) Extra "working" of material makes subsequent extrusion easier.
(4) Removal of moisture and volatiles.
(5) Improved powder flow properties.

Guimon (42) states that the output rates of twin screw extruders are directly proportional to the bulk density of the dry blend. In the preparation of dry blend; as the temperature increases, stabilisers and lubricants are absorbed by the PVC particles in the same way as plasticisers, and heat stability is better than those gently blended. By heating to temperatures over 100°C, moisture and other volatiles are eliminated which avoids defects such as voids and bubbles occurring in the extrusion.

Possible advantages of not mixing to a high temperature have been given as:-

(1) Time saving - faster output rate.
(2) No cooler necessary.
(3) No possibility of degradation during mixing.
(4) Lower energy requirement.

Considering the advantages of increased bulk density and the ill defined term "extra working", this appears to assume a need to select conditions such that the subsequent physical properties approach those of granules. Higher bulk densities could improve economics by giving higher output rates for a given screw speed. Additionally a higher bulk density could reduce the risk of frictional overheating by enabling lower screw speeds to be used while producing the same output rate.

It can be seen from the graphs that the degree of increase in bulk density will depend on the polymer (Fig. 24) and lubricants being used (Fig. 25-27). In some cases small changes in lubricant will have significant effects, in others they will not. The desirability of increasing bulk density will also depend on the effect of lubricant on fusion rate.

Although a complete examination of the effects of discharge temperature on absorption of stabiliser has so far not been carried out, an examination of electron micrographs of sectioned PVC particles has so far shown no evidence of absorption of solid lead stabilisers. A typical example is shown (Photograph 1.).

Effect of mixer discharge temperature on colour

For comparative purposes the formulation and materials were the same as those used in work previous to these investigations.

PVC Polymer	100
Tribasic lead sulphate	3
Glycerol monostearate	1

Four polymers were used. Mixing was carried out in a Papenmeier TLHK8 (8 litres) high speed mixer, rotor speed 3000 rpm without jacking cooling (see Appendix 9. 1). The temperature was recorded using a thermocouple consisting of spot welded wires protruding from a probe made of electrical braiding impregnated with epoxy resin, as this was more sensitive than the one fitted by the manufacturer. Small samples of powder were extracted using a special sampling device which was inserted through the hinged inspection port on the lid of the mixer. 4g of each sample were then pressed into discs in a hydraulic press with heating limited to three minutes at 115°C. Light reflectance of the disc was measured using an EEL spectrophotometer fitted with a No. 601 blue filter which gave a measure of the degree of pink colour developed as a result of thermal degradation. As hydrogen chloride splits off, conjugated double

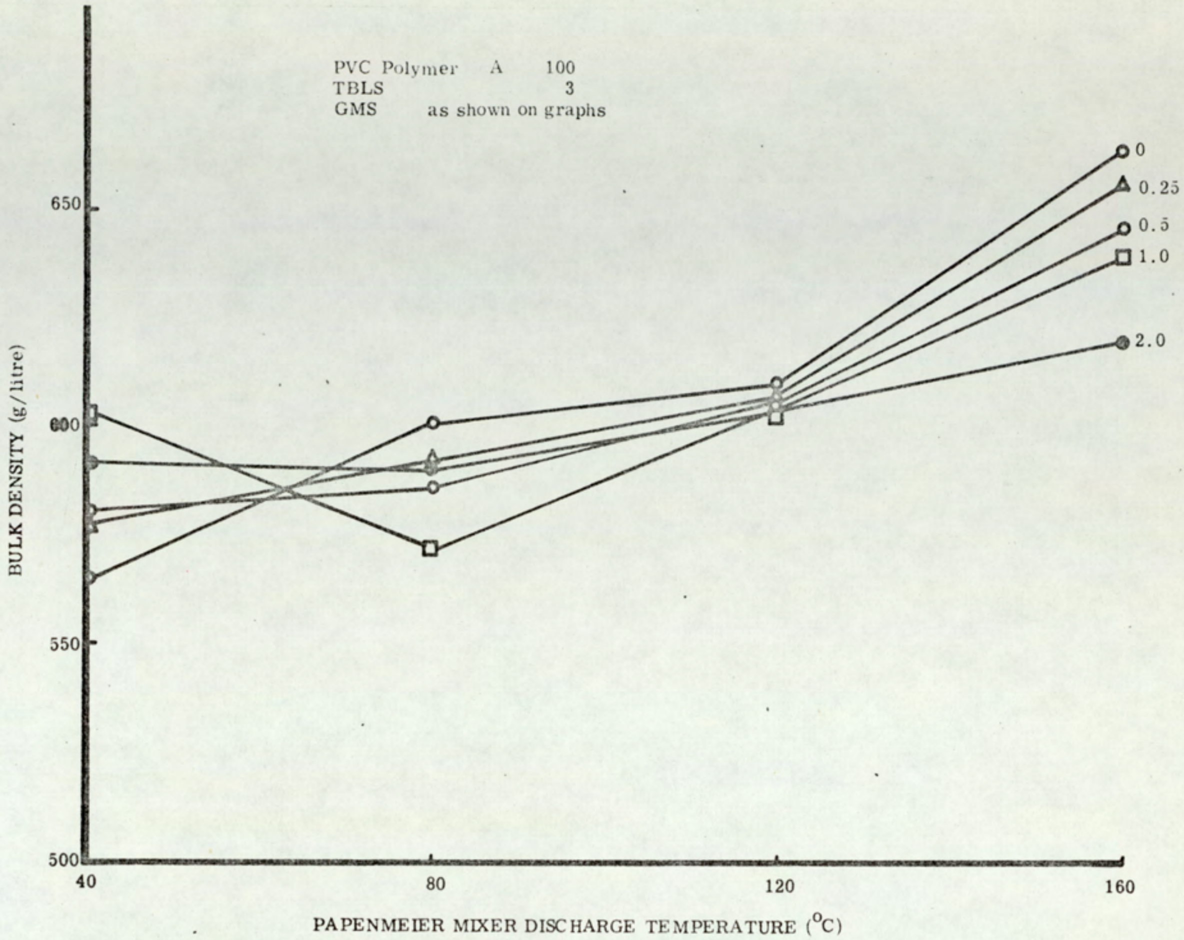


Figure 25 - Change in bulk density with mixer discharge temperature - Polymer A

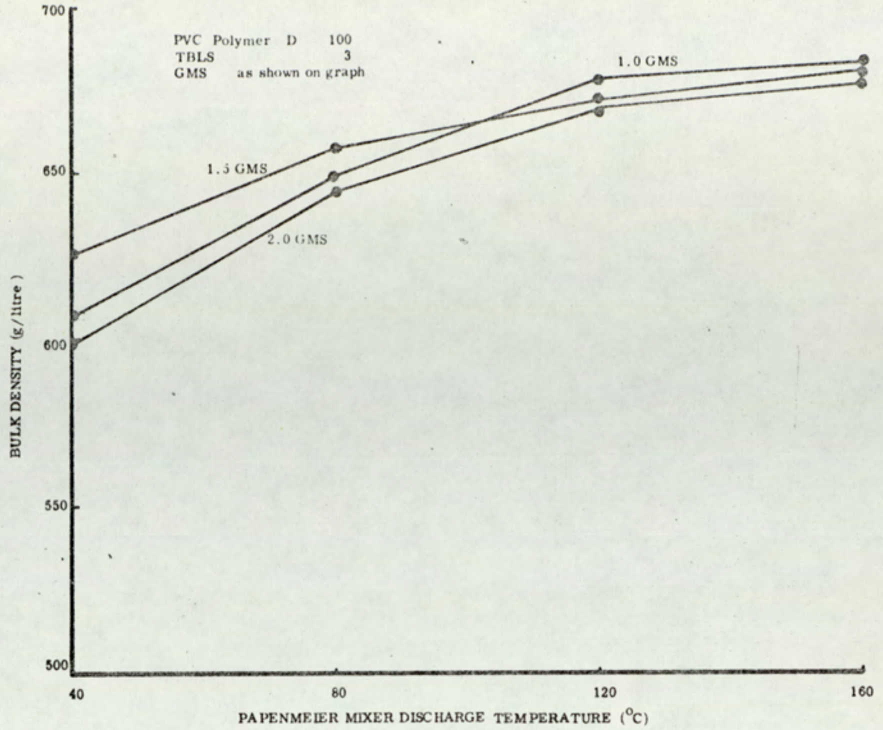


Figure 26 - Change in bulk density with mixer discharge temperature - Polymer D

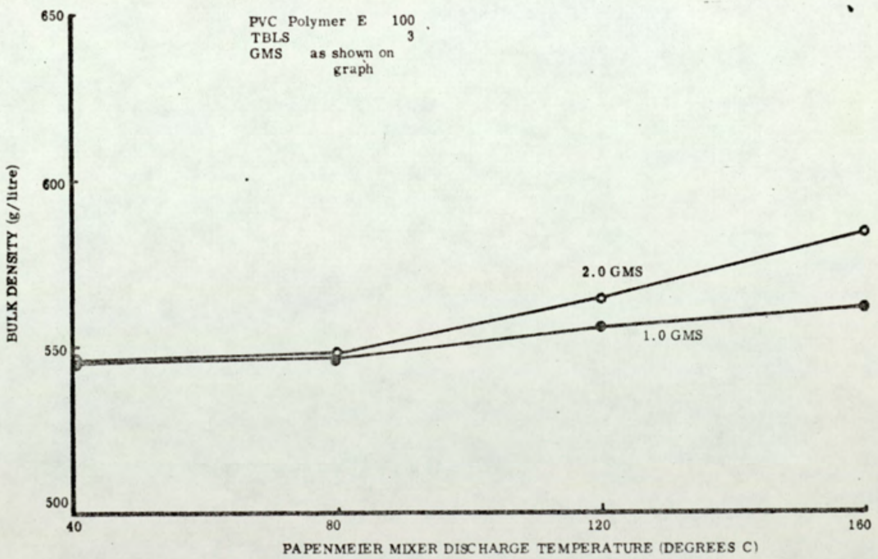


Figure 27 - Change in bulk density with mixer discharge temperature - Polymer E

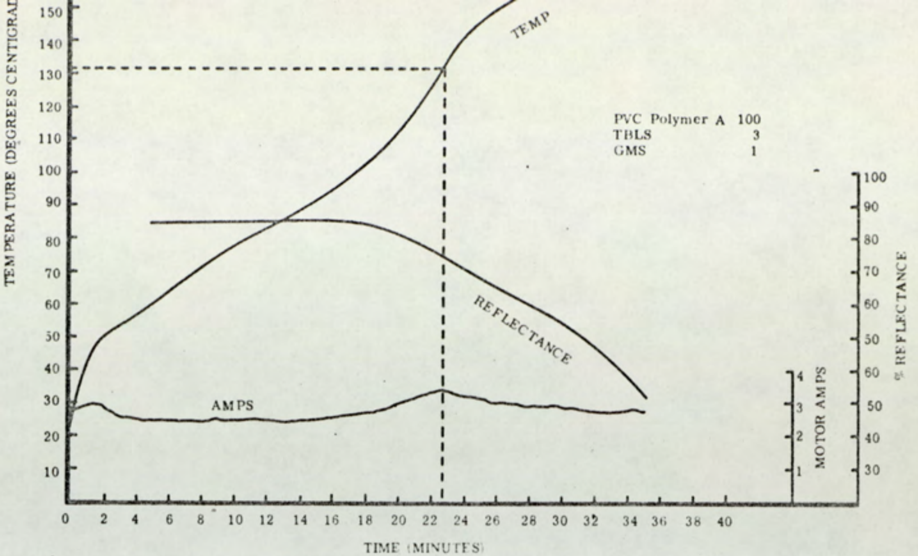


Figure 28 - Change in temperature, colour and motor power during high speed mixing Polymer A

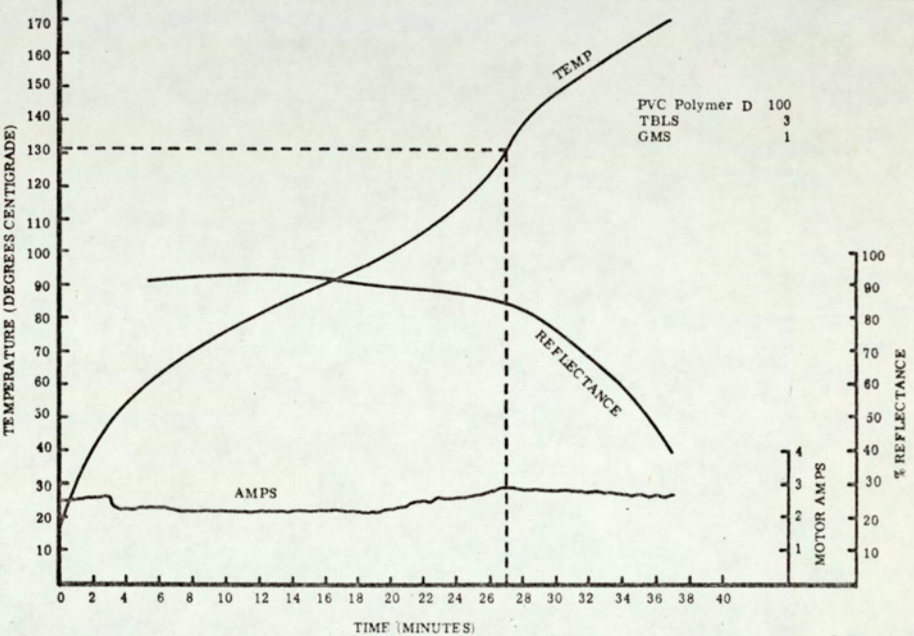


Figure 29 - Change in temperature, colour and motor power during high speed mixing - Polymer D

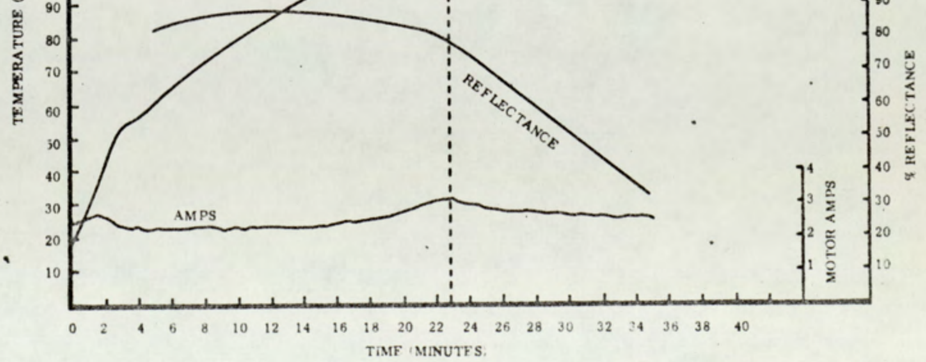


Figure 30 - Change in temperature, colour and motor power during high speed mixing Polymer E.

bonds are formed, seven consecutive units being sufficient to give a visually pink colour. At 80% reflectance the polymer sample appeared white while at 40% it appeared a pronounced pink.

Figs. 28-30 show curves for temperature, colour change (reflectance) and motor power consumption (amps) plotted against time during mixing. The temperature rises rapidly at first until the lubricant melts and then rises more slowly with a corresponding fall in motor current. At about 100°C the rate of temperature rise and power consumption increased, reaching a peak at 130-135°C in most cases. Above 140°C the rate of temperature rise was much lower and the motor current decreased.

The colour was fairly constant up to about 90-100°C. The improvement which sometimes occurred between 60°C and 80°C was attributed to improvements in the dispersion of lead stabiliser by the melting action of the lubricant when it melted. The solid opaque lead stabiliser had a pigmenting action. At temperatures above 140°C the colour changed to pink, becoming pink/brown at 170° - 180°C accompanied by evolution of hydrogen chloride. Change in colour with temperature is shown separately in Fig. 31.

Five PVC suspension polymers from two suppliers were used having K values (dichlorethane solvent) between 57 and 60, and graphs are shown for three representative grades.

Polymer A had a knobbly particle which was opaque.
" D had a more spherical particle which was also opaque.
" E had a more spikey but slightly glossy particle.

4.3 Effect of mixer discharge temperature on bulk density

Figs. 25-27 show changes of bulk density with high speed mixer discharge temperature for different levels of GMS as lubricant with polymers A, D and E. It was expected that increases in bulk density would occur with increasing discharge temperature.

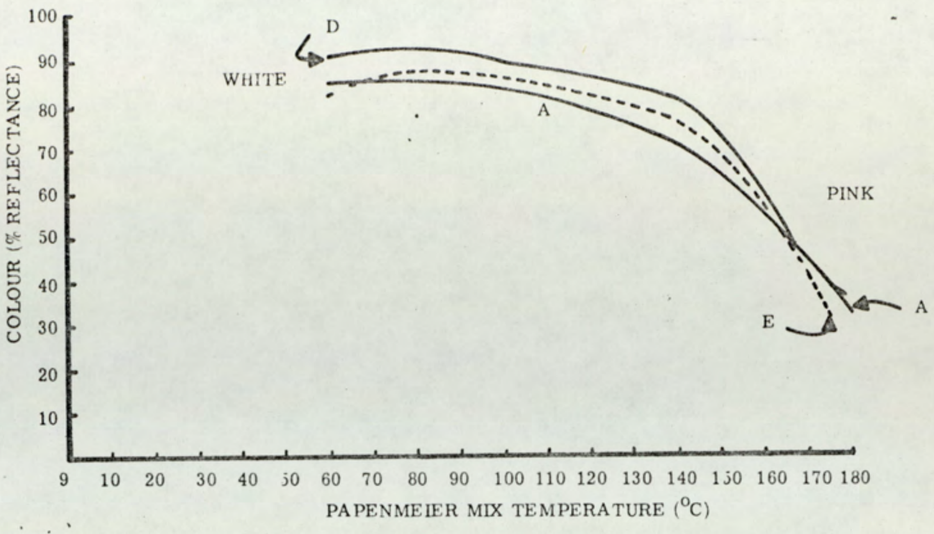


Figure 31 - Development of pink colour during mixing - comparison of polymers

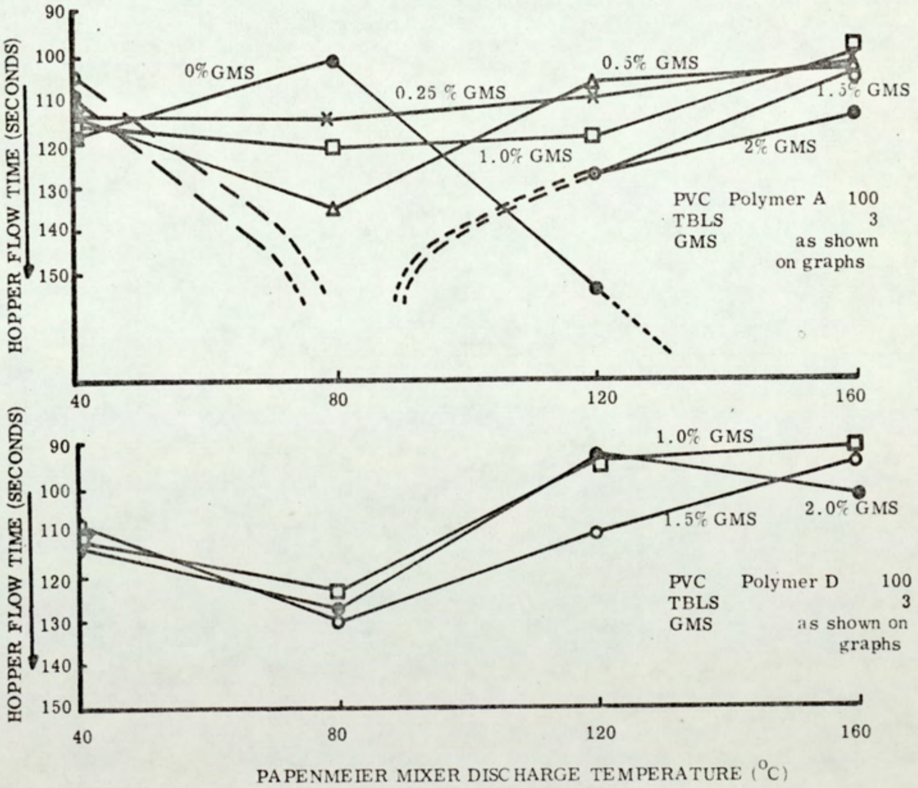


Figure 32 - Change in hopper flow time with high speed mixer discharge temperature

The increase could be caused by collapse of voids within the particles, by removal of roughness peaks and by increased packing as a result of surface changes due to additives. Depending on the nature of the polymer the bulk density/discharge characteristics varied. Thus the more spherical, higher packing particles of polymer D gave rapid increases in bulk density over the range 80-120°C while polymer A increased mainly over the range 120-140°C. Polymer E with the more glassy particles showed only slight increases in bulk density.

4.4 Effect of mixer discharge temperature on particle flow properties

'Hopper flow' tests were carried out to ASTM, D1895-65T where the time was measured for a given quantity of powder to run out of a small stainless steel funnel of specified dimensions. (see appendix). Results are shown for polymers A and D (Fig. 32) where it car be seen that GMS tended to form a cohesive surface at temperatures immediately above its melting point of 65°C, but at higher temperatures, where softening of the PVC took place, absorption of GMS was probably occurring to give improved flow properties. As the level of GMS was raised, this effect became more pronounced. Polymer D with its more spherical particles can be seen to have exhibited superior hopper flow results to polymer A. Figure 33 shows that if calcium stearate (which is a solid powder over this temperature range), was used as lubricant, then the problem of cohesion between 60 and 100°C was avoided.

4.5 Effect due to lubricant on bulk density and hopper flow for dryblends discharged at 120°C

A correct balance of lubrication for extrusion can be achieved by using mixtures of GMS and stearic acid as explained elsewhere. The effect of various ratios making a total of 2 phr lubricant on bulk density and on hopper flow are shown in Figure 34

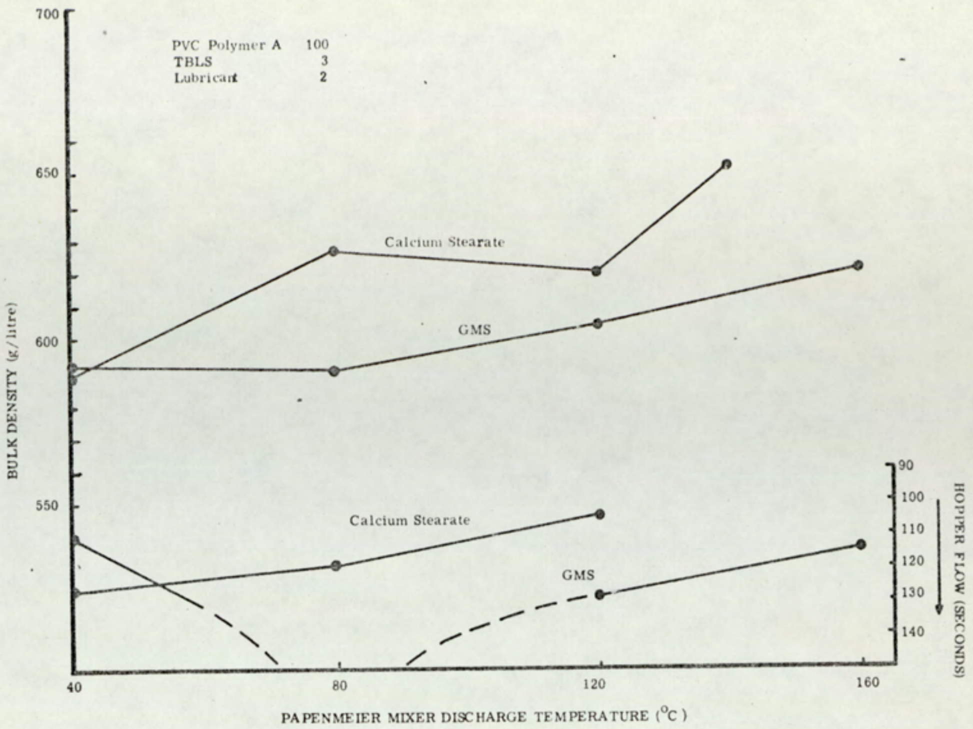


Figure 33 - Change in bulk density and hopper flow with high speed mixer discharge temperature - effect of lubricant

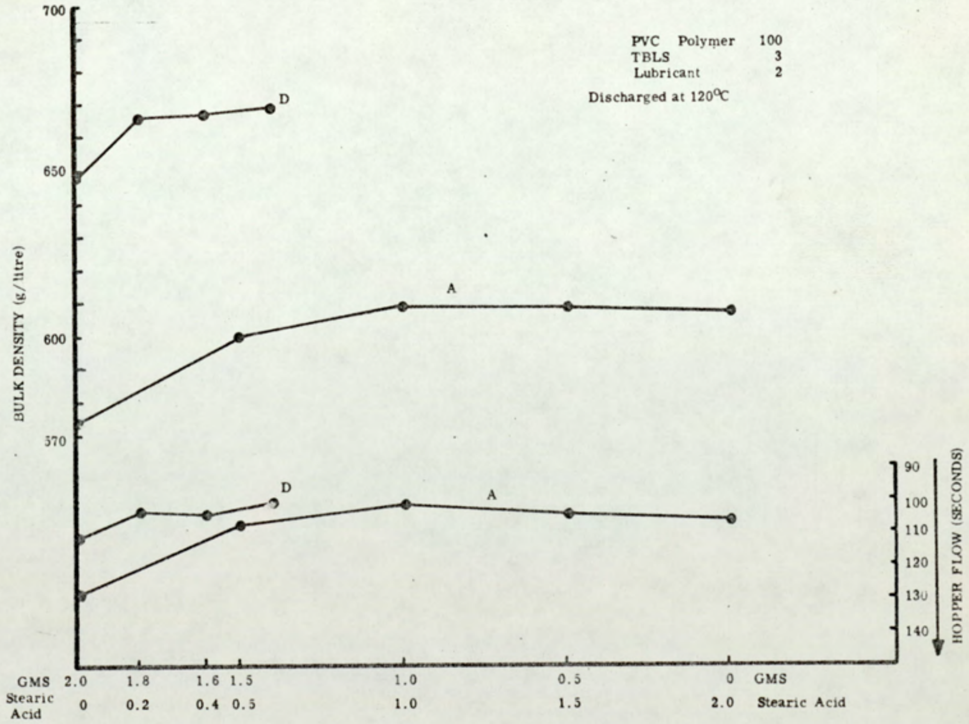


Figure 34 - Influence of lubricant on hopper flow properties

It can be seen that the bulk density and hopper flow effects are related and that these properties are improved by the external lubricant, even at relatively low concentrations.

Effect of mixer discharge temperature on sieve analysis

Sieve analysis was carried out for a wide range of mixes and it was expected that changes in bulk density and hopper flow properties would be associated with changes in sieve analysis data. However, differences due to either lubricants or mixing conditions were, in general, very small. Three examples are tabulated. The only significant changes were the shift from the 150 to the 100 mesh for polymer E at 160°C and the slight increases in proportion retained by the 100 mesh sieve of dry blends for polymer A with 1 and 2 pts GMS as lubricant and discharged from the mixer at 120°C.

Effect of mixer discharge temperature on extrusion

Dry blends were made up with the following formulation and discharged at 40, 80, 120 and 160°C.

PVC Polymer	100
Tribasic lead sulphate	3
Glycerol monostearate	2

They were then extruded from powder on a $1\frac{1}{2}$" single screw extruder described in the appendix. The dies used were a 7.9 min (5/16") rod and a channel profile as described in the appendix. Screw speeds were 20 and 30 rpm respectively, the higher speed being possible with the channel as it had a relatively thin section. The extruded rods were notched and impact tested on a Hounsfield "Charpy" type pendulum impact tester (see appendix), while the channels were impact tested on a falling weight machine.

The extruded channel was marked every minute during

TABLE 4 Sieve analysis:- Polymer A

PVC Polymer A	100
TBLS	3
GMS	2

		Percentage retained on sieve								
Sieve Mesh No.		60	72	85	100	150	200	240	300	Fines
Papenmeier mixer discharge temperature	40 C	½	1	3½	32½	46	12	2	1½	1
	80	0	1	3½	30	49½	12½	2	1	½
	120	0	1	3½	36	44	12	2	1	½
	160	0	1	3½	31	47½	13	2	1	1

TABLE 5 Sieve analysis:- Polymer D

PVC Polymer D	100
TBLS	3
GMS	2

		Percentage retained on sieve								
Sieve Mesh No.		60	72	85	100	150	200	240	300	Fines
Papenmeier mixer discharge temperature	40 C	0	1	2½	28½	47	15	3	2	1
	80	0	1	2½	27½	50	13	3	2	1
	120	0	½	2	24	53	14	3	2	1½
	160	0	½	1½	22	53	15	3	2½	2½
	172½	0	½	1½	21	53½	17	3	2	1½

TABLE 6 Sieve analysis:- Polymer E

PVC Polymer E	100
TBLS	3
GMS	2

		Percentage retained on sieve								
Sieve Mesh No.		60	72	85	100	150	200	240	300	Fines
Papenmeier mixer discharge temperature	40 C	½	1	2	18½	49	19½	4	2½	3
	80	0	1	1	12	54½	19	5	2½	5
	120	0	½	1	12	57½	20	3½	3	3
	160	0	½	2	29½	47	16	3	1½	½

extrusion and subsequently cut and weighed. The mean output rate and standard deviation were calculated for each run.

Although the screw compression ratio for these experiments was 3.0:1, the results for polymer E cannot be compared directly with those for A and D as the screw used for this run was a replacement which although nominally the same was in fact slight different.

The results in Tables 7-9 of output rate for different high speed mixer discharge temperatures were less variable for the channel extrusion than for the rod, due to the effect of higher die pressure for the channel extrusion (1300 psi for channel; 1000 psi for rod). Results at 80°C mixer discharge temperature were low in both cases and so erratic as a result of hopper bridging that data could not be obtained for standard deviation. The reason for this can be seen in figure 32 where 80°C discharge materials were shown to have poor flow properties. The relatively high standard deviation at 120°C discharge temperature was also due to powder flow being poor although it was much improved over that at 80°C. With polymer A (Table 7), the effect of poor powder flow at 80°C was still evident, particularly with the rod extrusion, but was not as serious (with the channel) as polymer E. Impact strength of the rod from material discharged at 40°C was considerably higher than values for the other discharge temperatures and could be associated with the presence of unfused material as described later.

With polymer D, (table 8) output rates for the channel extrusion were very consistent with little variability for a particular discharge temperature dryblend and between different dryblends. Rod extrusion output rates varied between different discharge temperature dryblends and could not be correlated with

TABLE 7

PVC Polymer A

	Channel 30 rpm				Rod 20 rpm	
Discharge Temperature	Bulk Density	Average O/P rate	Standard Deviation	Impact Strength	O/P rate	Impact Strength
	g/l	g/min		Kgf. cm	g/min	Kgf. cm
40 C	592	155	3.9	9.1	133	13.8
80	590	156	5.8	8.3	116	7.6
120	604	156½	4.7	8.6	134	8.7
160	620	160	5.8	6.9	152	7.8

TABLE 8

PVC Polymer D

	Channel 30 rpm				Rod 20 rpm	
Discharge Temperature	Bulk Density	Average O/P rate	Standard Deviation	Impact Strength	O/P rate	Impact Strength
	g/l	g/min		Kgf. cm	g/min	Kgf. cm
40 C	599	164	1.9	9.4	119	-
80	645	164	1.4	7.2	141	6.5
120	670	161	2.7	8.3	139	7.8
160	677	166	2.0	6.9	131	6.8

TABLE 9

PVC Polymer E

	Channel 30 rpm					Rod 20 rpm	
Discharge Temperature	Bulk Density	Average O/P rate	Standard Deviation	Impact Strength	Colour	O/P rate	Impact Strength
	g/l	g/min		Kgf. cm		g/min	Kgf. cm
40 C	545	151	5.63	5.4	57	117	-
80	548	135	-	5.4	54	102	10.0
120	565	160	18.9	6.1	54	106	8.3
160	584	168	8.75	6.9	56	123	8.0

measured dryblend properties. As polymer D had a high bulk density and more spherical particles, powder flow properties were improved and fusion would have been earlier, which would explain the consistent output rates for the channel die. The variations in output for the rod die can be associated with the lower head pressure.

In general these results show that poor hopper flow properties will give variations in output rate, the amount of variation being dependent upon the die pressure. In practice such faults could be rectified by changing the mixer discharge temperature in accordance with the requirements as shown in the previous graphs. This would apply if the alternative of increasing the level of incompatible lubricant was unacceptable. (e.g. where transparency is important).

4.8 Effect of mixer discharge temperature on extruder fusion rate

The idealised process of direct powder extrusion can be summarised as follows:- The dryblend is prepared in a high speed mixer and consists of particles of PVC polymer coated with lead stabiliser, lubricant and pigment. This powder is then fed into the hopper of the extruder. The powder is free flowing and is removed from the hopper throat at a continuous even rate by the screw and conveyed along the barrel through the feed (or transport) zone. The material is then heated and compressed in the compression (or melting) zone and is fully fused at the beginning of the metering zone. This zone, feeds the die with a homogeneous material of uniform temperature where it is shaped into the required profile. The PVC compound will contain a suitable type and level of lubricant to prevent sticking to metal parts of the machine and to impart a good surface finish to the extrudate.

It appears that in practice the process is not so simple (14) The extruder output rate and the quality of the extruded pipe will depend on the properties of the material in the fully gelled state which will be effected by the fusion behaviour, which in turn will be effected by the powder properties. Comparing this type of extrusion with conventional extrusion of material in granule form (e.g. 3 mm cubes), the extruder has the extra function of converting the PVC powder into a uniform melt. This part of the process cannot be dealt with separately as any change which influences fusion rate may influence the melt properties and hence the finished product quality.

The shear forces bearing on the PVC particle will also depend on the bulk density of the powder. This in turn is influenced by the high speed mixing conditions and particularly on the discharge temperature.

It was, therefore, considered likely that variables resulting from changes in mixer discharge temperature such as lubricant adsorption bulk density and interparticulate friction would effect fusion rate in the extruder screw. A torque rheometer can be used to measure lubrication effects on fusion rate but this was found to be unsuitable if bulk density differences existed. For this investigation the extruder was fitted with a screw extraction system so that the fusion rate could be measured directly from material removed from the screw channel.

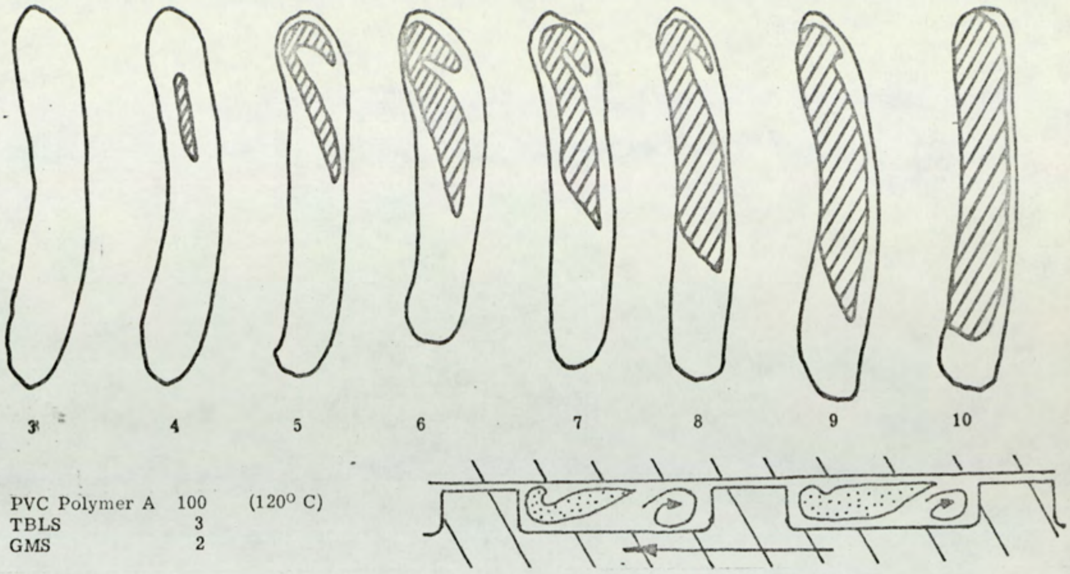
For this work the extruder was fitted with a 7.9 mm (5/16") dia rod die. Extrusion was carried out in the normal manner and the extruded PVC rods were later cut into 32 mm ($1\frac{1}{4}$") lengths, notched and tested on a Hounsfield Impact tester ("Charpy" type). When the last of the batch of PVC powder was about to be conveyed from the hopper, the extruder motor was stopped and the die removed from the end of the barrel. The hydraulic hand pump was then operated and the screw forced out. A line was drawn with a marker axially along the length of the screw starting from flight termination, and the melt was numbered at every revolution of the flight. The PVC was then unwound from the screw channel and laid out flat until cool. The strip was then cut on the marked line giving a numbered length from the screw channel representing a sample for every revolution of the flight. The face was then wet sanded on progressively finer abrasive paper and finally polished using "Belco" rubbing compound. A 3 mm wide strip containing the polished face was then sawn off and the sample mounted under a low powered microscope using the method described in the following section and illustrated in the appendix. The outlines of the sections and the unfused areas were traced on paper using an "Abbe camera lucida", which was clipped to the eyepiece. This had a prism arrangement which allowed the drawing being made (and the pencil tip) to be seen as a second image superimposed on the microscope image. From the drawings, measurements of the total area and area of fused material were made and a figure for percentage fusion was obtained from the ratio of these two areas. The boundaries were normally distinct enough as unfused polymer was discoloured pink, while dispersed lead stabiliser in the fused area pigmented the section white or pale cream. Oven heating was used where necessary to darken the unfused pink area by further degradation.

The cross sections during fusion consisted of an area of sintered powder surrounded by an enveloping layer of fused (or melted) PVC with a melt pool at the rear of the channel as described by Maddock (54). As the fusion progressed, the melt pool (at the rear of the channel) became progressively larger at the expense of powder and cross channel circulating flow appeared to deform the sintered bed of powder into a hook configuration. By plotting fusion by the area ratio method described against distance along the screw, fusion rate curves were obtained as shown. This meant that the fusion rate could be characterised under certain given extrusion conditions of temperature screw speed and screw compression ratio.

Figure 34 shows tracings of the cross sections of the PVC in the screw channel as seen under the microscope at a very low magnification. The outlines of the sections and pink unfused areas are shown shaded. The shapes varied due to distortion during removal from the screw channel. For this particular dryblend formulation, with polymer A, 100% fusion was achieved 3-4 turns from the screw tip. At a point 10 turns from the screw tip, only about 25% fusion had occurred. The rate of fusion is shown in figure 36. Figure 37 shows drawings of sections of a similar dryblend mixed and extruded under the same conditions but in this case the PVC was polymer D. This polymer fused more rapidly than polymer A under these conditions, probably due to its higher powder bulk density, otherwise the pattern of fusion was similar to that for A. Rate of fusion is shown in figure 38.

Experiments were confined to a screw of 2:1 compression ratio as that of 3:1 was unavailable at the time, but the small differences in fusion rate due to bulk density differences would be even smaller with a high compression screw. It was concluded that changes in fusion rate as a result of using polymers of different bulk density were relatively small, while feasibility studies on this apparatus concerning the effects of lubricants showed them to have very significant effects, so efforts were then concentrated on this aspect of extrusion.

As discussed earlier, the choice of a lubrication system in PV powder extrusion is a variable which concerns the converter and one in which there are both the problems of evaluating the very large number



PVC Polymer A 100 (120° C)
TBLS 3
GMS 2

Figure 35 - Cross sections of material removed from screw channel - Polymer A

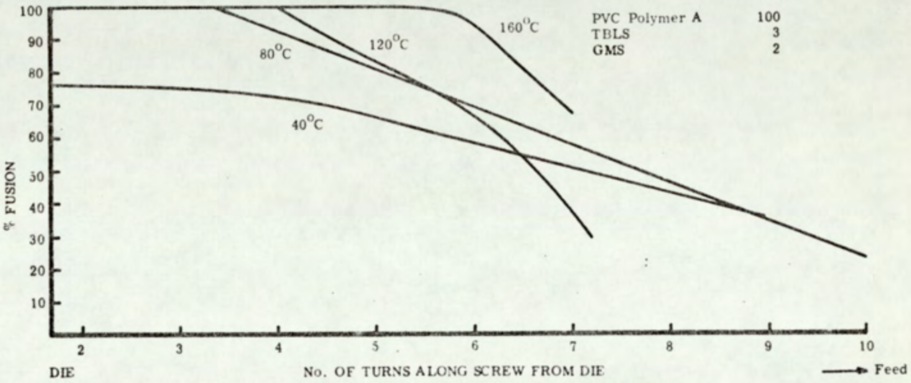
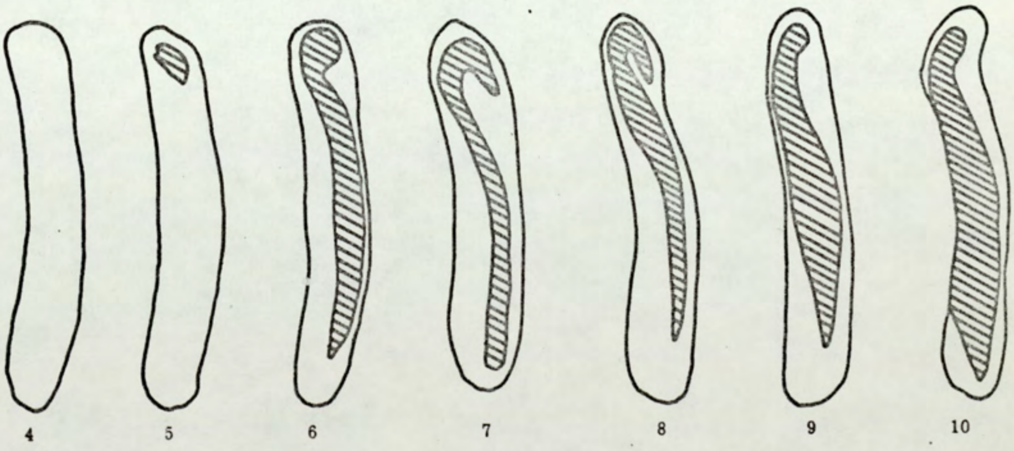


Figure 36 - Effect of mixer discharge temperature on fusion rate - Polymer D



PVC Polymer D 100 (120°C)
TBLS 3
GMS 2

Figure 37 - Cross section of material removed from screw channel - Polymer D

available (far more than grades of suitable polymer) as well as that of determining the optimum combination and level. As discussed earlier, the effects could well be very large in proportion to the amount used. A further complication was that the only published data at that time on fusion rate measurements in a single screw extruder concluded that the fusion mechanism was different for PVC powder to that of other polymers and that even with extremes of temperature and shear bordering on thermal degradation, only 90% fusion could be achieved, yet the above experiments indicated a fusion mechanism not very different to that of Maddock and Street, while 100% fusion was also achieved.

It was established by microscopic examination that the appearance of the sections shown in figures 35 and 37 (occurring when GMS was used as lubricant) was a result of a fusion mechanism similar to that described by Street, where a melt pool collected at the rear of the channel. The sections shown in fig. 39 (occurring when substantial levels of "incompatible" lubricants such as stearic acid were used) were the result of the mechanism described by Menges and Klenk (8, 9) where material is fused only where wedged between flight land and barrel surface. In this case the melt pool collects at the forward side of the channel.

For descrimination purposes these mechanisms will be termed "rear (of the channel) fusion" and "forward fusion" respectively. It can be seen from graphs 36, 38 and 40 that the "rear fusion" mechanism is relatively rapid, whereas the "forward fusion" mechanisms is very slow and probably incapable of achieving complete fusion of the powder.

These mechanisms were investigated later in some detail (see section 5. 7).

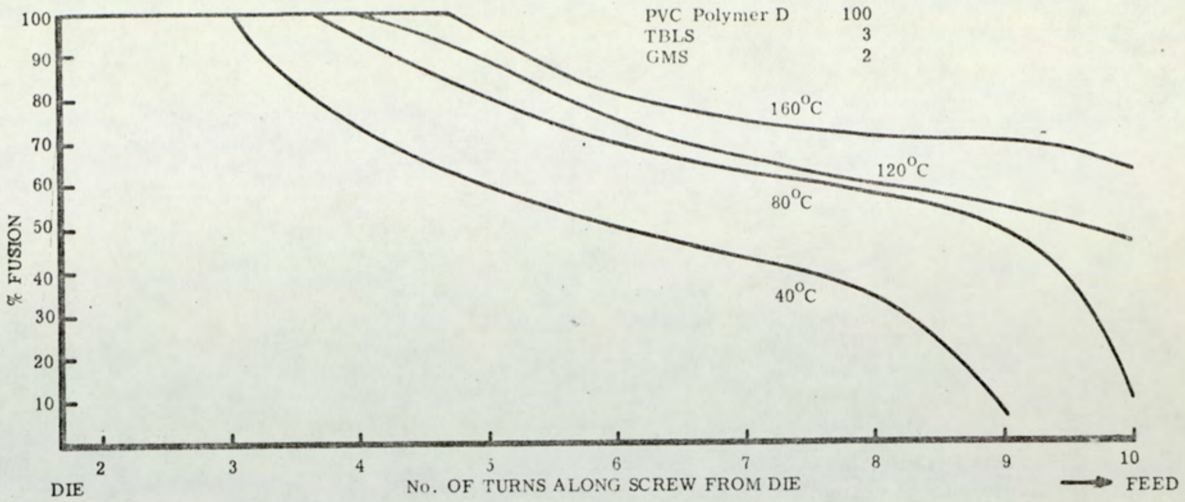
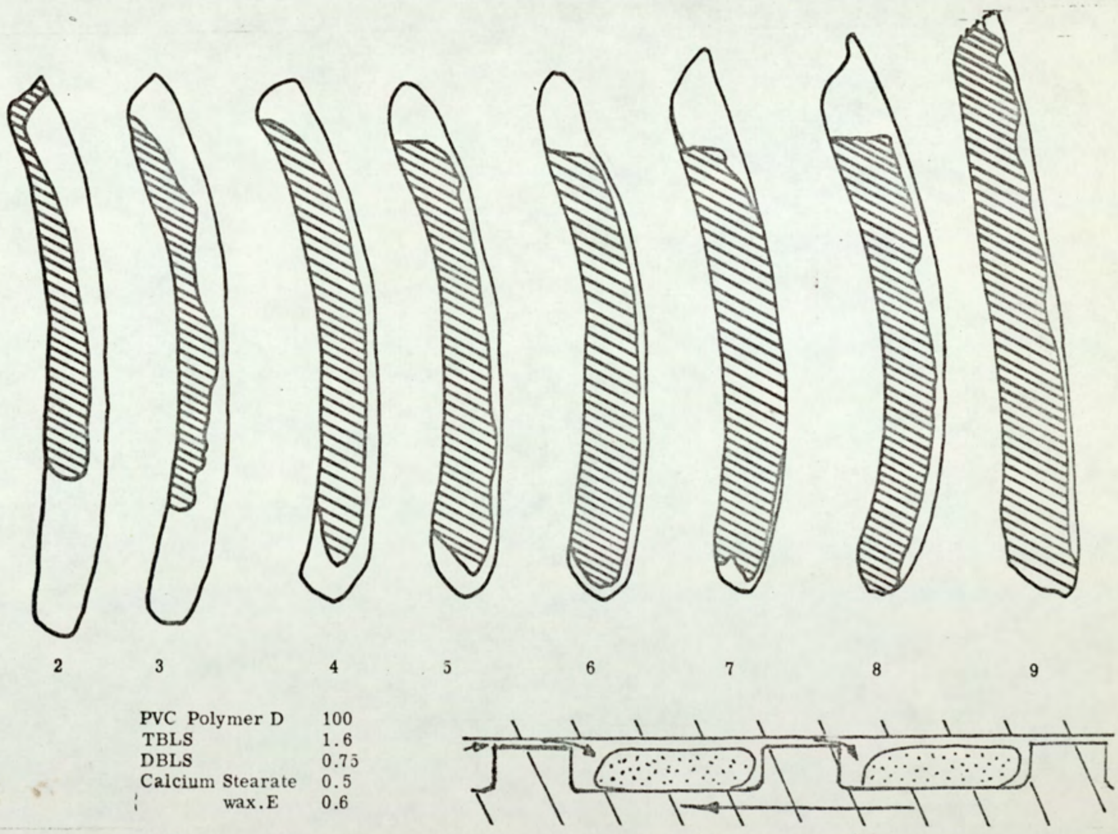


Figure 38 - Effect of mixer discharge temperature on fusion rate - Polymer D



PVC Polymer D	100
TBLS	1.6
DBLS	0.75
Calcium Stearate	0.5
wax.E	0.6

Figure 39 - Cross sections of material removed from screw channel - "Commercial type" formulation.

5. Behaviour of various compositions during extrusion

5.1 Examination of the effects of the relative proportions of "compatible" to "incompatible" lubricant on fusion rate

From Fig. 39 showing drawings of the sections, it appeared that the amount of incompatible (or external lubricant) was too high for the extruder and fusion rate was too slow to achieve 100% fusion by the time the screw tip was reached. Total fusion at this point was only 65%. Comparing these sections with those described above, it appeared that the outlines of the sintered powder were different and that shear forces resulting from cross channel circulation had caused this material to slip across the channel during the last two turns. The rate of fusion for this material is shown in Fig. 40. The extruded rod had a matt finish, a knobbly appearance and was pink in colour. By contrast the formulation containing 2.5 phr G M S as lubricant (shown in fig. 40 fused relatively quickly and produced a smooth glossy rod, white in colour.

The number of materials available as lubricants for unplasticised PVC is extensive and the problem exists as to how representative experiments can be carried out to evaluate the effects of lubricants in general without using a very wide range

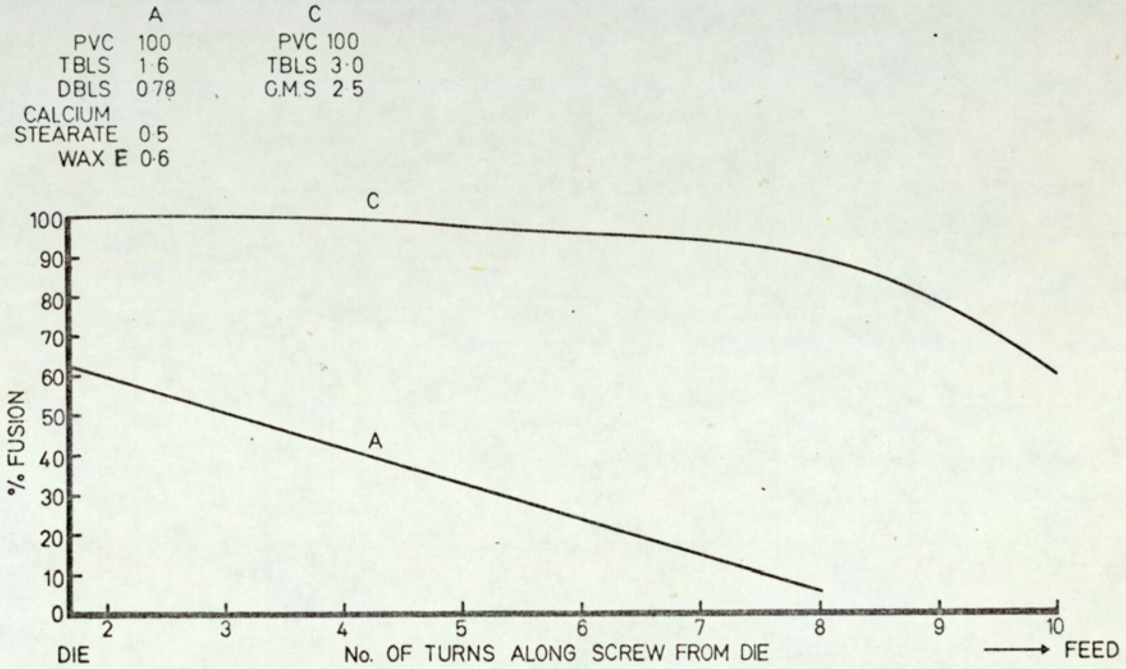


Fig. 40 Effect of formulations on fusion rate

in numerous combinations at various levels of addition. These difficulties have been described in a paper by Oakes & Hughes

where an extensive examination of a wide range of lubricants was carried out using a Brabender Plastograph. Jones & Hawkes (14) have described a procedure for determining suitable lubrication properties including control of fusion rate using a mixture of different lubricant types. According to Bursian (61) "external" lubricants could often be entirely dispensed with when the quantity of "internal" lubricant was kept sufficiently high or if the lubricant used was dual functional. High levels of internal lubricant could considerably depress the PVC softening point, but they were indispensible and when external lubricants were used, internal ones should always be combined with them.

For these investigations a mixture of "compatible" (internal) and "incompatible" (external lubricants was used) and in order to achieve a wide range of lubrication properties, the two types were varied in their relative proportions in a similar manner to mixtures of normal lead stearate and dibasic lead stearate described by Jones & Hawkes (14). G M S was used as the compatible lubricant as it had been used in previous experiments and results of fusion behaviour in the screw channel and in the Brabender Plastograph showed that increasing level of GMS had only small effects on fusion rate. The extrusion results for 2.5 phr GMS used alone as lubricant agrees with Bursian's statement concerning "internal" lubricants. Similar experiments showed that very small increases in stearic acid concentration caused relatively large decreases in fusion rate. This was also shown by Jones & Hawkes (14). (A further reason was that GMS and Stearic acid are available in similar physical forms and have similar melting points at about 60°C).

The range examined was as shown in Table 10

The results are shown in Fig. 41. Using 2.5 and 2.0 pts GMS, 100% fusion was reached well before the screw tip. By replacing 0.5 pts GMS with stearic acid the fusion was delayed to an extent that only 80% fusion was achieved at the screw tip. Further replacement of GMS by stearic acid caused total fusion to be further reduced so that at stearic acid levels of 1.0 phr and above the material was substantially unfused.

Figures 42 and 43 show rates of fusion for dry blends of polymer D with a lubricant system consisting of a mixture of GMS

TABLE 10

PVC Polymer A	100	100	100	100	100	100
TBLS	3	3	3	3	3	3
GMS	2.5	2.0	1.5	1.0	0.5	0
Stearic Acid	0	0	0.5	1.0	1.5	2.0

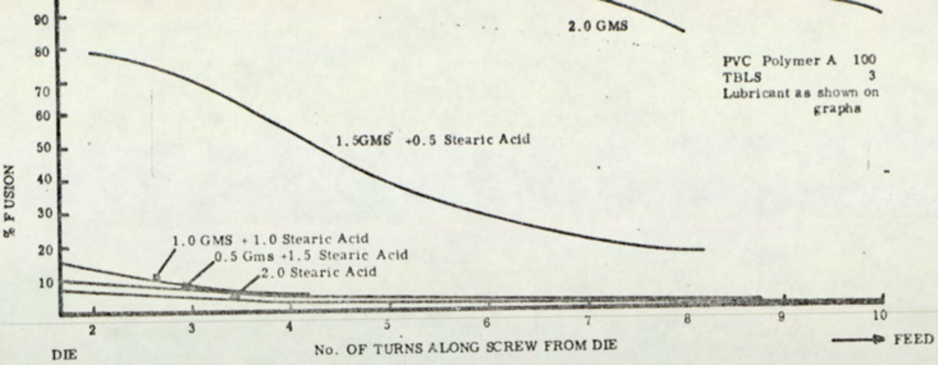
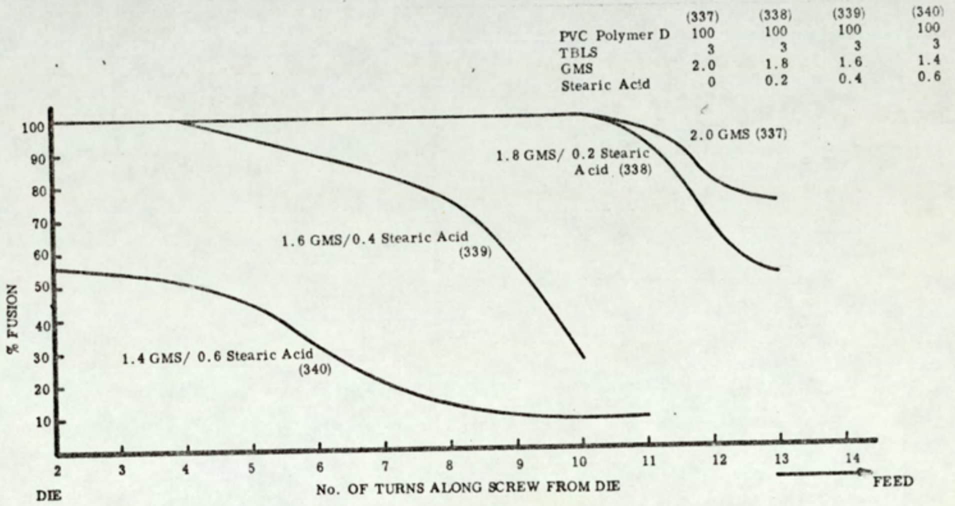
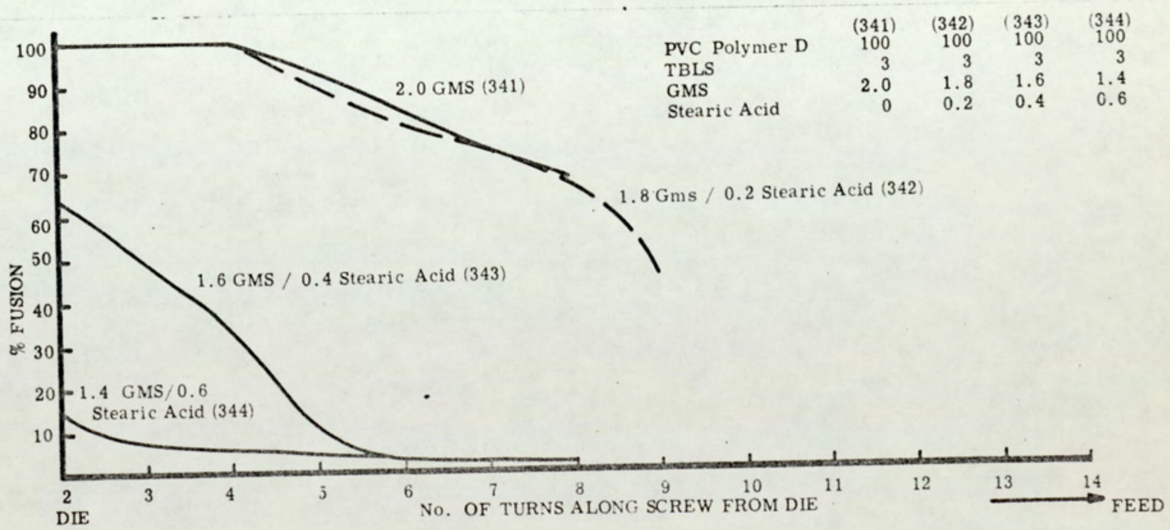


Figure 41 - Effect of lubricant on fusion rate - Polymer A with 3:1 depth ratio screw



	(337)	(338)	(339)	(340)
PVC Polymer D	100	100	100	100
TBLS	3	3	3	3
GMS	2.0	1.8	1.6	1.4
Stearic Acid	0	0.2	0.4	0.6

Figure 42 - Effect of lubricant on fusion rate - Polymer D with 3:1 depth ratio screw



	(341)	(342)	(343)	(344)
PVC Polymer D	100	100	100	100
TBLS	3	3	3	3
GMS	2.0	1.8	1.6	1.4
Stearic Acid	0	0.2	0.4	0.6

Figure 43 - Effect of lubricant on fusion rate - Polymer D with 2:1, depth ratio screw

and stearic acid similar to those in Figure 41 but in this case the stearic acid increments were reduced to 0.2 and the range limited to 1.4 pts GMS/0.6 pts stearic acid. Comparisons were also made between screws having compression ratios of 2.0 and 3.0:1. The relative positions of the curves in both graphs are almost identical. If displaced, those on Figure 43 can be superimposed on Figure 42 with the screw tip starting at $8\frac{1}{2}$ turns from the die. 0.2 phr stearic acid had very little effect on fusion rate, but above this level, very small additions had significant effects, 0.4 being excessive for the 2:1 screw and 0.6 being excessive for the 3:1 screw.

5. Examination of individual lubricant effects in a commercial type of formulation.

The work was continued more systematically with improved instrumentation of the extruder to examine in more detail the contribution of a number of lubricants in a typical commercial formulation. A similar formulation was used to one described in commercial literature (62). The level of each lubricant was varied in turn in suitable increments and the effects measured using fusion rate measurement techniques previous described. Although parallel studies on correlation of extruder fusion rates with Brabender Plastograph fusion times showed many anomalies (which were explained in later investigations) it was shown that efficiency of the lubricant depended on its compatibility in the formulation as a whole and on the wetting nature of other additives. Thus lubricants should be evaluated as part of a complete formulation of stabiliser, other lubricants, process acids, etc., similar to ones in which they are to be used.

The effects of concentration changes were expected to provide information on the overall effectiveness of the lubricant and provide data for lubricant adjustments to satisfy requirements of die changes, and extruder changes where screws may differ. A contact in industry commenting on these results considered that many attempts to solve a lubrication problem were prolonged by the use of alternative lubricants that probably behaved similarly to the one being replaced.

At the same time as the Wax E evaluation, comparisons were made with similar formulations extruded conventionally from fused granules so that possible requirements of lubricant changes as a result of 'change-over' from granule to powder feed could be determined.

5.1 Effect of Wax E

The first lubricant examined was Wax E (a modified Montan Wax) which is commonly used in unplasticised PVC formulation as an "external" lubricant. The experiments were carried out on dryblend formulations (shown in table 11) prepared in a high speed mixer and discharged into a cooler at 120^{o}C.

The dryblends were extruded with a 1½" dia single screw extruder having a 24:1 C/D barrel with either a 2.0:1 or 3.0:1 compression screw and fitted with an 8 mm (5/16") dia rod die containing a melt thermocouple and a melt pressure transducer. A constant screw speed of 30 rpm was used. The screw jacking system and fusion rate measurement were the same as before. In the tables the fusion point number is the number of turns from the screw tip at which the material reached 100% fusion, i.e. it gives the number of turns of the channel containing 100% fused material. When the percentage fusion at the screw tip was less than 100, the value reached at this point has been recorded. With this series of formulations a number of samples were obtained in which the melt contained partly dispersed patches of unfused particles after the rolling melt had reached the full width of the channel. As the amounts were small and they fused within 2-4 turns they were not included in fusion rate measurements.

The results in table 12 and Fig. 44 show that increasing the level of Wax E from 0.5 to 0.7 phr had only small effects on

TABLE 11

PVC (suspension polymer)	100	100	100	100	100	100
Tribasic lead sulphate (damped)	2	2	2	2	2	2
Disbasic lead stearate (damped)	0.5	0.5	0.5	0.5	0.5	0.5
Calcium stearate (damped)	0.5	0.5	0.5	0.5	0.5	0.5
Wax E	0.5	0.6	0.7	0.8	0.9	1.0

fusion rate and other extrusion effects, but a further increase of 0.1 phr to 0.8 phr decreased the fusion rate until a point was reached where the material was barely fused at the screw tip.

At 0.5-0.7 phr the fusion mechanism involved a melt pool at the rear of the channel as described by Maddock (54) and by Street (53). At 0.8 phr this fusion model still predominated but there was evidence in the region of the metering zone that a melt pool collected at the forward side of the channel as described by Menges and Klenk (9.10). A further increase of Wax E to 0.9 and 1.0 phr resulted in a complete change to the forward fusion model and incomplete fusion of the PVC. This condition is in agreement with the findings of Menges and Klerk and is discussed in detail later.

Output rate increased with decreasing fusion rate as a result of increasing lubricant level. In terms of production control, the output rate sensitivity meant that strict control of lubricant level is essential. In this particular case an increase in Wax E level of 0.1 phr could have had serious effect on quality, while smaller changes would have been sufficient to send extrudate dimensions outside specified limits.

The effects of screw compression ratio and die land length are shown in Table 13 and Fig. (45) using a Wax E concentration of 0.6 phr.

Effect of screw compression ratio and die land length

The over all effect was that where the compound was not overlubricated, the rate of fusion was determined by the screw compression ratio and hardly effected by the die land length over the range used. The effect of the die was more important with the lower compression screw as fusion was only just or not quite complete and therefore influenced by die back pressure effects.

Although the 2:1 compression screw was supplied for PVC granule extrusion, it had been previously used for powder with some lubricants. With the formulation described having an 0.8 phr Wax E level,fusion failed to reach 100% with either the 1:1 or 9:1 land length dies. With the 3:1 screw, complete fusion was only achieved with the 9:1 die only. The output rates were again higher for the lower rate of fusion and were overall proportionally lower for the 2:1 screw which had a shallower feed zone. The very low degree of fusion with the short die also gave a reduction in melt pressure.

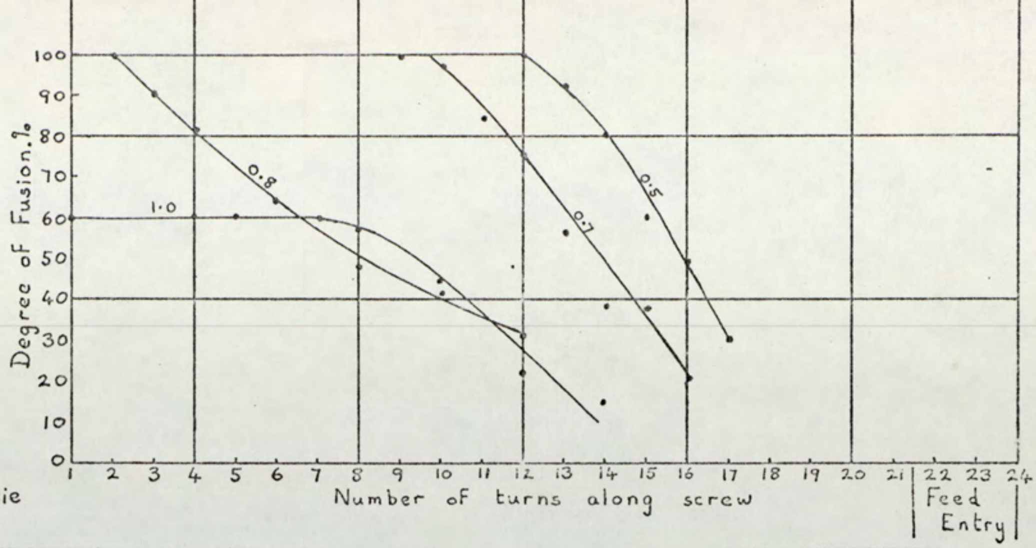


Figure 44 - Fusion rate - Effect wax E concentration

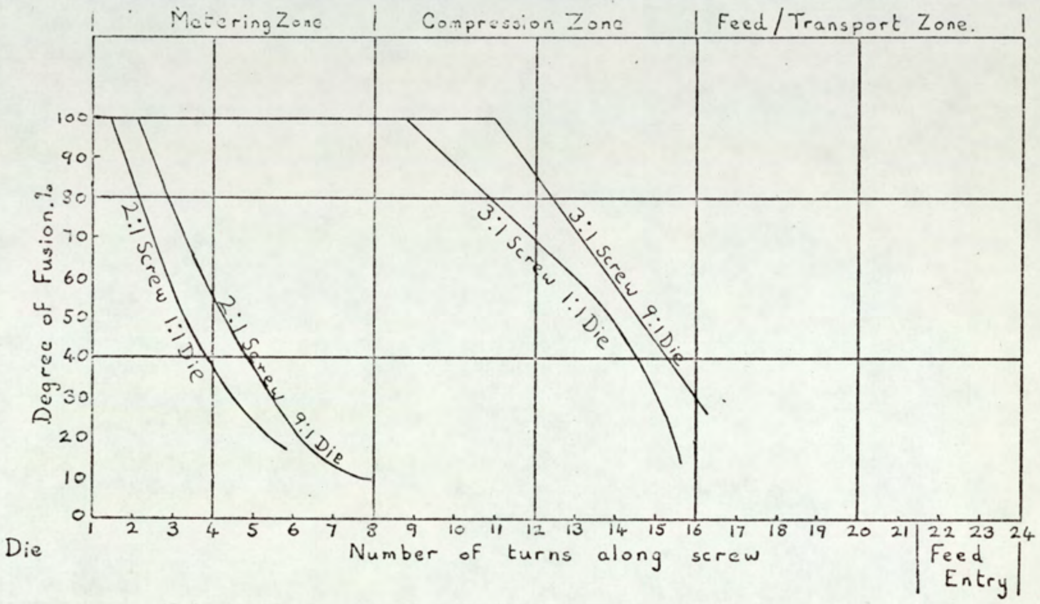


Figure 45 - Fusion rate - Effect of screw compression ratio and die land length ratio

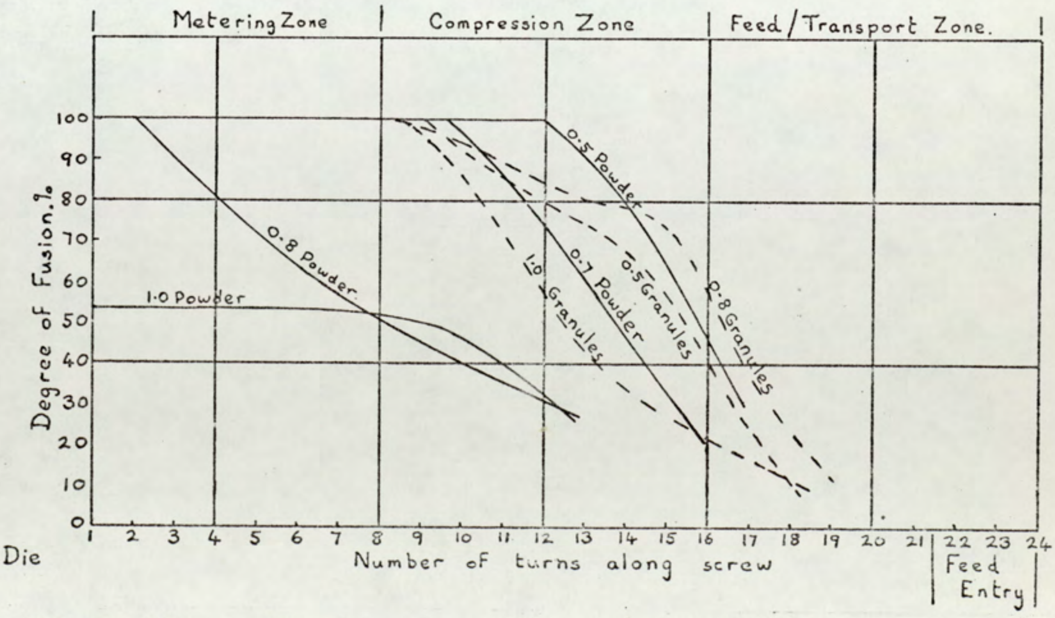


Figure 46 - Fusion rate - comparison of powder with granules

TABLE 12 *Effect of concentration of wax E*

WAX E CONTENT	DRIVE CURRENT	DIE MELT PRESSURE	DIE MELT TEMP.	FUSION MODEL	FUSION POINT	OUTPUT RATE
phr	A	lbf/in^2	C			g/m
0.5	9½	1900	200	Rear	12	161
0.6	10½	2500	192	Rear	11	184
0.7	10½	2325	191	Rear	10	199
0.8	9½	2500	185	*Rear	2	231
0.9	8	2350	182	Forward	(95%)	226
1.0	7¼	3240	203	Forward	(60%)	231

*Both models occurred simultaneously, but rear fusion predominated

TABLE 13 *Effects of screw compression ratio and die land length at two lubricant levels*

WAX E CONCN.	SCREW COMP. RATIO	DIE LAND LENGTH RATIO	DRIVE CURRENT	DIE MELT PRESSURE	FUSION MODEL	FUSION POINT	OUTPUT RATE
phr			A	lbf/in^2			g/m
0.8	2.5:1	9:1	9½	2500	Rear	2	231
	2.5:1	1:1	9¼	2700	Forward	(100%)	240
	1.5:1	9:1	6	3250	Forward	(50%)	180
	1.5:1	1:1	6	2575	Forward	(25%)	217
0.6	2.5:1	9:1	10½	2500	Rear	11	184
	2.5:1	1:1	10½	700	Rear	9	185
	1.5:1	9:1	6	2750	Rear	2	178
	1.5:1	1:1	5¾	1250	Rear	1	180

TABLE 14 *Comparison of powder feed with granule feed*

WAX E CONTENT	FEED	DRIVE CURRENT	DIE MELT PRESSURE	DIE MELT TEMP.	FUSION MODEL	FUSION POINT	OUTPUT RATE
phr		A	lbf/in^2	C			g/m
0.5	Powder	9½	1900	200	Rear	12	161
0.5	Granules	10¼	1975	193	Rear	8	187
0.8	Powder	9½	2500	185	Rear	2	231
0.8	Granules	10¼	1950	192	Rear	7	182
0.9	Powder	8	2350	182	Forward	(95%)	226
1.0	Powder	7¼	3240	203	Forward	(60%)	231
1.0	Granules	10½	2250	195	Rear	6	191

At 0.6 phr Wax E, the effect of screw compression ratio was dominant and although rear fusion behaviour was obtained with both screws, a compression ratio of 2:1 was obviously inadequate. For production control purposes these results illustrate the problem of using the same formulations on varying combinations of extruders and dies.

Table (14) and Figure (46) illustrate the differences between powder and granule extrusion and account for the good performance shown by single screw extruders in the survey described earlier and their decline in recent years for this application. The powder blends were prepared in duplicate, and one batch was extruded as powder, the other as granules. The granules were prepared by dividing the dryblend,one half being milled with a small pigment addition, the other being milled unpigmented. The material was removed from the mill in strips about 75 mm wide and 3 mm thick and granulated into cubes. The coloured and unpigmented granules were tumbled together and then extruded, the coloured granules giving a clearer identification of fusion pattern. As a result of an indistinct boundary between melt and unmelted granules the percentage fusion was based on the relative proportions of melt rolling melt width and not on relative areas.

The results show that a rear fusion model was obtained with the lubricant content ranging from 0.5 to 1.0 phr becoming 100% fused at 9 to 13 turns respectively from the screw tip. As already described, the powder was only just fused at 0.8 phr Wax E and was incompletely fused at 0.9 and 1.0 phr. Another feature of the granule extrusion was the relatively small change in output rate with increasing Wax E content. Die pressures were more consistent, remaining differences being probably due only to lubrication effects in the die exist region.

5.3 Effect of dibasic lead stearate and calcium stearate

These two materials have been grouped together, as being metals soaps they are dual functional, acting both as stabiliser and lubricant. They also differ from waxes, fatty acid esters etc. in that their melting points are higher than normal PVC processing temperatures.

Dibasic lead stearate is used primarily as a heat stabiliser although being a stearate it has some lubricating effect. According to Jones and Hawkes (14), lubrication of a compound can be controlled by varying the ratio of dibasic lead stearate to normal lead stearate which is a more effective lubricant (and a less effective stabiliser).

Calcium stearate is used primarily as a lubricant, but being a hydrogen chloride "acceptor", also contributes to the heat stability of the compound, although its stabilising efficiency is relatively small. Its widespread use commercially is mainly due to its relatively low cost.

A range of 0, 0.5, 1.0 and 1.5 phr was used for each lubricant in combination with Wax E similar to that described previously.

Dryblends were made up in an 8 litre high speed mixer and discharged into a cooler at 120°C. The extrusion conditions and results are shown in tables (15) and (16), and the fusion rates in Figs. (47) and (48).

In general the results were very similar for the two lubricants. The fusion rates were very similar, as were the observed fusion patterns. At the point where fusion started the rear fusion mechanism occurred for all levels of lubricant, being followed by an undetermined mechanism showing trends towards forward or rear fusion patterns. The main difference was that the boundaries between melt and unfused material

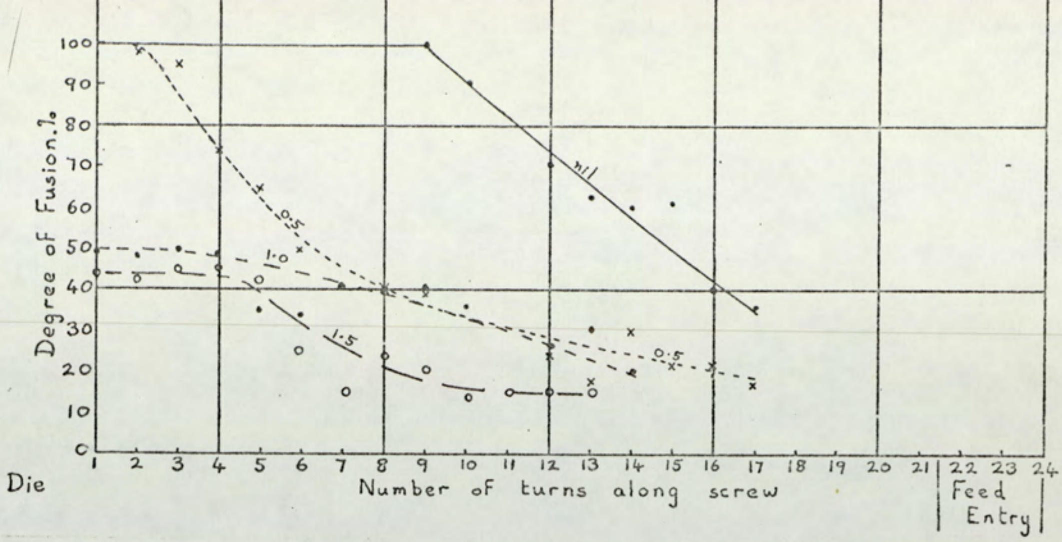


Figure 47 - Fusion rate - Effect of dibasic lead stearate concentration

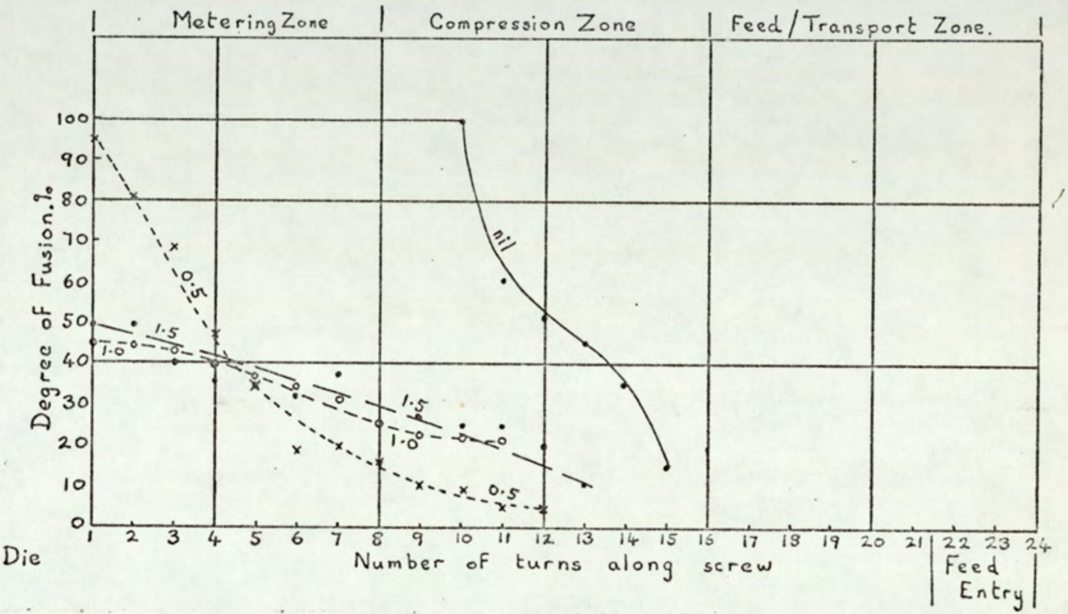


Figure 48 - Fusion rate - Effect of calcium stearate concentration

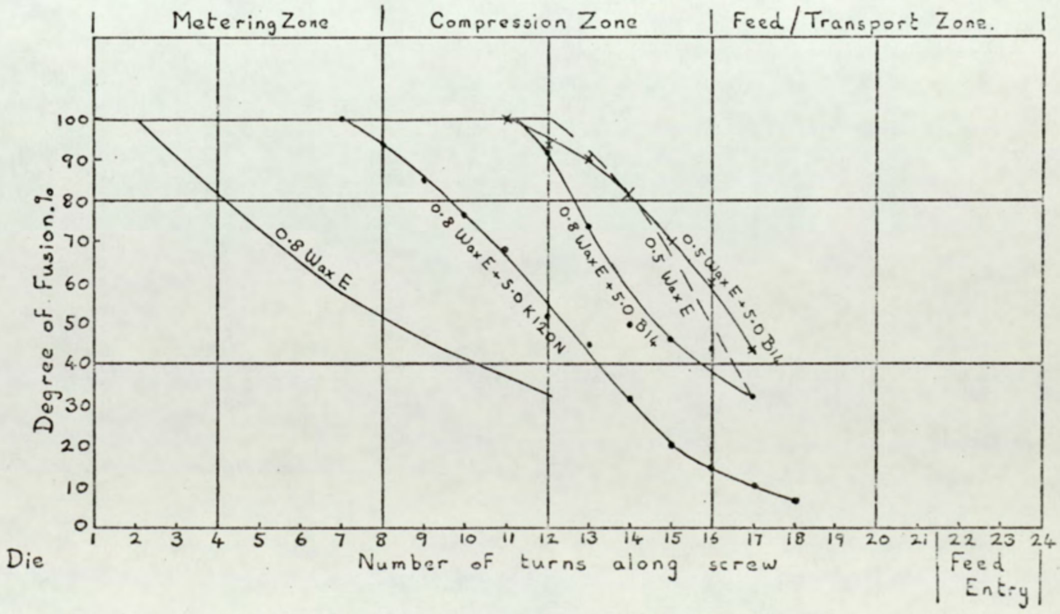


Figure 49 - Fusion rate - Effect of process aid and impact modifier

TABLE 15

Effect of Dibasic lead stearate

Expt No.	Level of DBLS	Temperatures °C					Melt press. psi	Screw speed rpm	Motor Current amp	Fusion pt.	O/P rate g/min	Appearance of extrudate
		B_1	B_2	B_3	Die	Melt						
1162	0	151	181	193	190	186	1600	30	$9\frac{3}{4}$	9	222	Smooth, dull surface
1163	0.5	139	175	187	191	182	2175	30	$8\frac{1}{2}$	1	242	Smooth glossy
1164	1.0	138	174	195	191	183	1775	30	$7\frac{3}{4}$	50%	234	Smooth wavey
1165	1.5	138	184	196	191	183	1800	30	$7\frac{3}{4}$	45%	254	Smooth wavey

TABLE 16

Effect of Calcium stearate

Expt No.	Level of Ca. St.	Temperatures °C					Melt press. (PSI)	Screw speed (rpm)	Motor current (amps)	Fusion pt.	O/P rate g/min	Appearance of extrudate
		B_1	B_2	B_3	Die	Melt						
1158	0	140	180	200	190	192	1300	30	$9\frac{1}{4}$	10	176	Rough
1159	0.5	131	169	193	190	188	1850	30	$6\frac{3}{4}$	95%	204	Rough
1160	1.0	133	168	189	190	196	2100	30	$6\frac{1}{2}$	45%	223	Glossy and wavey
1161	1.5	137	170	190	190	189	1250	30	6	50%	232	Glossy and wavey

were more clearly defined in the case of calcium stearate. The general shapes of the curves can be explained in terms of fusion mechanism. Once the overall lubrication level was such that the rear fusion mechanism did not predominate, fusion was incomplete and a further addition of lubricant had little further effect. Thus for both calcium stearate and DBLS, results for 1.0 and 1.5 phr levels were very similar. In the tables the trends are the same although some anomalies exist. In general, increasing the lubricant level decreased the fusion rate so that the temperatures developed from melt deformation became less, the motor power requirements were reduced and the output rate increased. In addition, the extrudate became more glossy and the tendency to become wavy increased.

Differences between calcium stearate and DBLS were that although fusion rates were very similar, DBLS gave a glossier surface than calcium stearate, while calcium stearate gave the higher output rate.

Effect of Process aids and Impact modifiers

A number of polymeric materials can be incorporated as blends into PVC to improve both processability and impact strength. These additives are normally used as powders which are incorporated into PVC dry blends, at levels of 5-10 phr. In this series the materials used were chlorinated polyethylene (CPE), acrylonitrile butadiene styrene copolymer (ABS) and methacrylate butadiene-styrene copolymer (MBS). MBS polymers used as impact modifiers have a higher butadiene content than the process aids.

The results are shown in Table (17) while the fusion rates for representative MBS systems are shown in Figure 49 The general effect is that these materials promote rear fusion so that at 0.5 Wax E level where fusion was already quite rapid, there was little effect on fusion rate, whereas when 0.8 phr Wax E was used these additives, even at a level of only 5 phr moved the point of complete fusion from the screw tip back to 10-12 turns so that far more mechanical work was applied to the material. This is further shown by the high melt temperatures and lower output rate. Although a wide range of materials were used the results were remarkably similar.

TABLE 17

Effect of polymeric modifiers

Wax E Content	Additive	Melt Temp °C	Moter Current Amps	Fusion Model	100% Fusion PC (Turns from die)	Output Rate g/min	Appearance of extruded rod
0.5	–	200	9½	Rear	13	161	Smooth slight gloss die lines
0.5	Acrylic Process Aid	192	10	Rear	12	161	Smooth medium gloss
0.5	Acrylic Impact Modifier (i)	205	9¾	Rear	12	152	Smooth mat
0.5	ABS Impact Modifier	207	10	Rear	10	155	Smooth mat
0.5	Chlorinated Polyethylene (i)	202	9	Rear	11	161	Smooth slight gloss die lines
0.5	Chlorinated Polyethylene (ii)	212	8¼	Rear	11	150	Smooth slight gloss die lines
0.5	Acrylic Impact Modifier (ii)	203	9¾	Rear	11	151	Even sharkskin
0.8	–	185	9½	Rear + some forward	2	231	Smooth very slight gloss
0.8	Acrylic Process Aid	195	10½	Rear	7	205	Smooth medium gloss
0.8	Acrylic Impact Modifier (i)	188	10¼	Rear	9	146	Smooth slight gloss
0.8	ABS Impact Modifier	190	10	Rear	11	170	Smooth medium gloss
0.8	Chlorinated Polyethylene (i)	190	8¾	Rear	12	162	Smooth matt/ sharkskin
0.8	Chlorinated Polyethylene (ii)	199	8¾	Rear	11	168	Smooth glossy
0.8	Acrylic Impact Modifier (ii)	182	10	Rear	11	160	Smooth medium gloss

Effect of filler

Stearate coated precipitated calcium carbonate (PCC) is now widely used as a filler in rigid PVC rainwater and soil pipes, and extruded profiles, both as a processing aid and to improve impact strength (48,49). The levels normally used are not high enough to impart very significant reductions in raw material cost. It has been shown that for maximum impact strength, an optimum filler level exists at about 7.5 phr for pipes made by direct powder extrusion on a twin screw machine (14).

Trials with a stearate coated PCC with DBLS, calcium stearate and Wax E as lubricants were made, but the melt filled screw could not be extracted from the barrel when the higher filler levels were used. Finally, after a number of trials, a typical low cost rainwater pipe formulation was used as follows:-

PVC suspension polymer Corvic D57/17	100
Tribasic lead sulphate	2
Calcium Stearate	1.0
Stearic Acid	0.4
Stearate coated PCC (Wynnofil S)	0-10

Dry blends were made up in the small high speed mixer with all ingredients added at the start. A deviation from this used by some

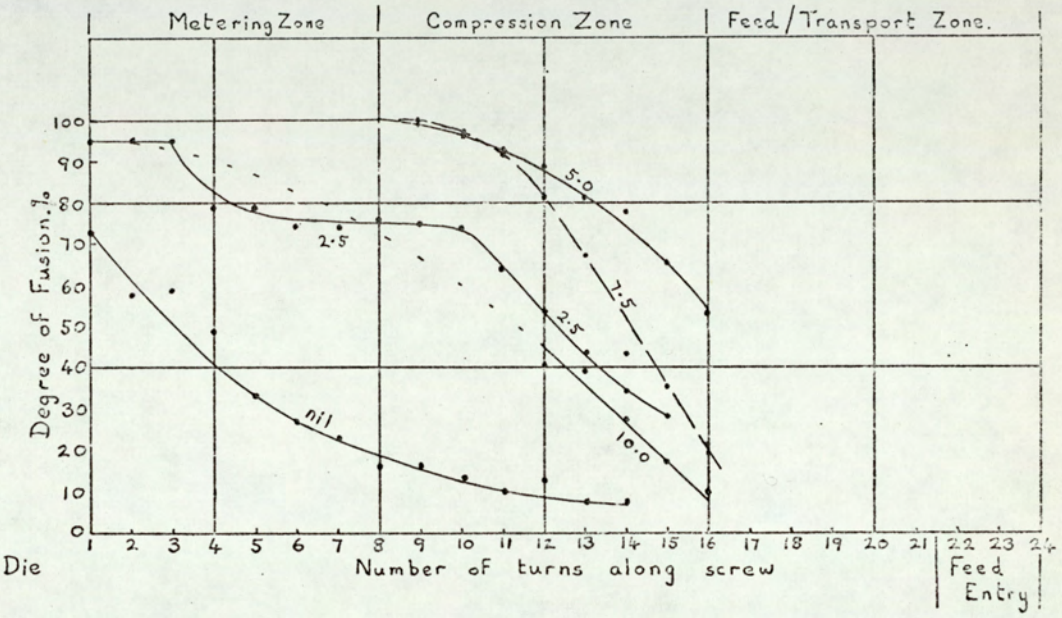


Figure 50 - Fusion rate - Effect of filler concentration

TABLE 18

		Temperatures											
Expt. No.	Level of PCC	B1	B2	B3	Die	temp °C	Melt press psi	Screw speed rpm	Motor amps	Fusion pt	O/P rate g/min	Impact Str ft/lbs	Appearance of extrudate
1166	Nil	134	174	189	194	174	2050	30	8	73%	255	.045	Glossy wavy
1167	2.5	148	179	189	190	176	1850	30	$8\frac{1}{2}$	94	227	.038	Glossy wavy
1168	5.0	155	182	195	191	180	1600	30	10	8	206	.040	Glossy smooth
1169	7.5	160	181	200	192	186	1500	30	$10\frac{1}{4}$	9	197	.038	Dull smooth
1170	10.0	160	180	190	195	187	1850	30	$10\frac{3}{4}$	-	178	.035	Rough surface

companies is to add the filler at the end of the hot mixing stage. The dry blends were discharged into a cooler at 120°C.

The extrusion fusion rates are shown in Figure 50.

As the fusion rate increases to give firstly a greater percentage fusion and then a greater length of screw containing melt (depending on the mechanism) then the temperatures increase, die pressures decrease, motor currents increase, and output rates decrease. The underfused extrudates were glossy due to high lubricant content, but wavy due to the formation in the conical die of a rod from a spiral strip from the screw channel containing a band of unfused material. When the material was fully fused, the extrudate was smooth, but at higher filler levels the surface gloss was lost, eventually becoming rough.

The results for 10 phr filler level do not completely fit the general pattern and this is further complicated by the failure to determine the degree of fusion for most of the screw length. This was due to the boundaries between melt and unfused material becoming ill-defined as a result of the presence of the filler, and discolouration through degradation. The higher melt pressure and the appearance of the material in the screw channel indicated that it had not quite fused, whereas barrel temperatures melt temperature motor current and output rate indicated a rapid fusion rate.

The graphs show that at zero filler level the lubrication level was excessive so that only 70% fusion was reached. At 2.5 phr, fusion level was raised to nearly 100%, while a further increase to 5.0 phr gave a fusion rate rapid enough to reach 100% at 8 turns from the screw tip, i.e. at the beginning of the metering zone. A further increase to 7.5 phr delayed onset of fusion but the rapid fusion rate resulted in complete fusion at 8 turns. At 10 phr onset of fusion was further delayed.

An optimum filler level appeared to exist when considering fusion rate and extrudate appearance. A possible explanation for its influence on fusion rate is as follows:-

The high surface area of the filler will be covered by a proportion of the stearic acid making less available for coating the PVC particles. The result is that fusion rate is increased. As fusion proceeds, then some of the "incompatible" stearic acid will tend to leave the surface of the filler and appear on the melt surface. This would account for the levelling off of the 2.5 phr fusion curve. At high levels of filler, the fusion rate will be reduced by the filler particles separating the PVC particles and preventing them from fusing. Other complicating factors are the lubricating effect of the calcium stearate on the surface of the filler and the increase in shear due to the volume effect of the filler, (i.e. effectively increasing the compression ratio).

Impact strength results do not show an optimum filler level. In previous work it was found that orientation resulting from "draw-down" (stretching while still "fluid") increased the impact strength of extruded rod. In this case, different die pressures and output rates require different amounts of draw down to achieve the required diameter, while the presence of unfused material in spiral strata may enhance impact strength (60). Other work using falling weight impact tests on extruded pipe did show an optimum filler level at about 7.5 phr.

5.6 Effect of screw speed on fusion rate.

A problem existed in obtaining stable temperature conditions within a reasonable running time. As the screw speed was raised, the amount of heat generated by shear deformation increased, so that the temperature of the barrel steadily increased during the experiment. Figs. 51-53 show that with 2.0 phr GMS as lubricant the fusion rate was decreased a small but significant amount by the increase in screw speed, but at 1.0 GMS/0.4 stearic acid (figs. 54-56) and 1.0 GMS/1.0 stearic acid (figs. 57 and 58) the screw speed increase had practically no effect. Overall it appears that the main influence on fusion rate is the lubricant and not the screw speed.

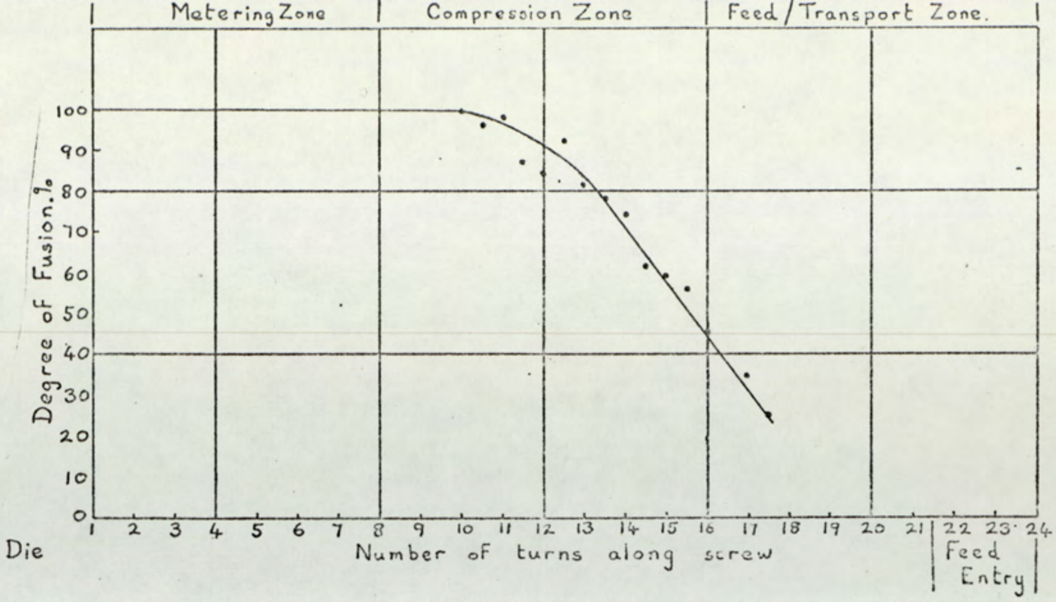


Fig. 51 Fusion rate - Lubricant: GMS 2.0/stearic acid nil. Screw speed 30 rpm.

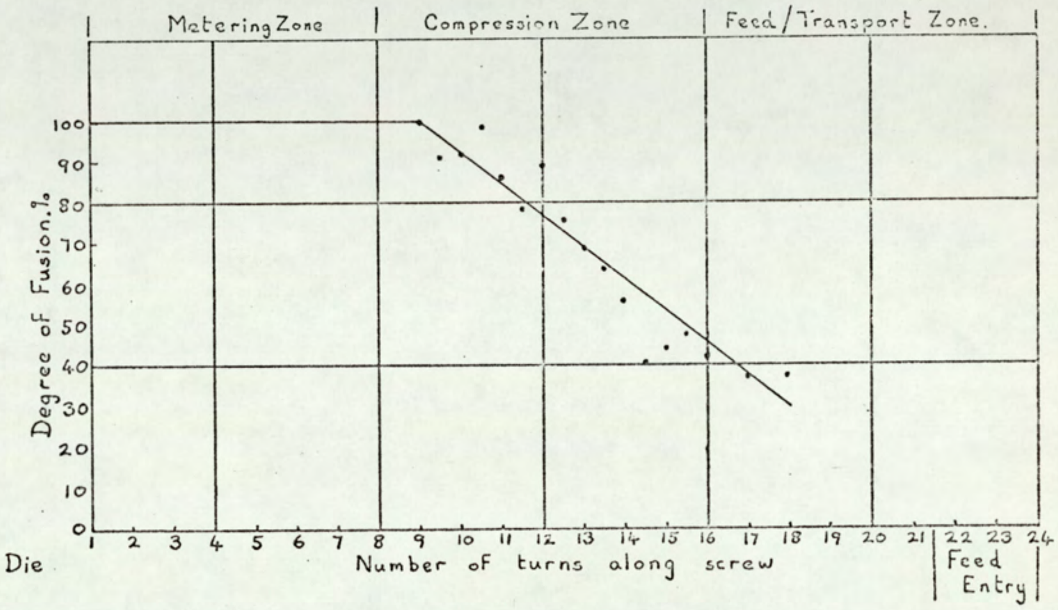


Fig. 52 Fusion rate - Lubricant: GMS 2.0/stearic acid nil. Screw speed 50 rpm

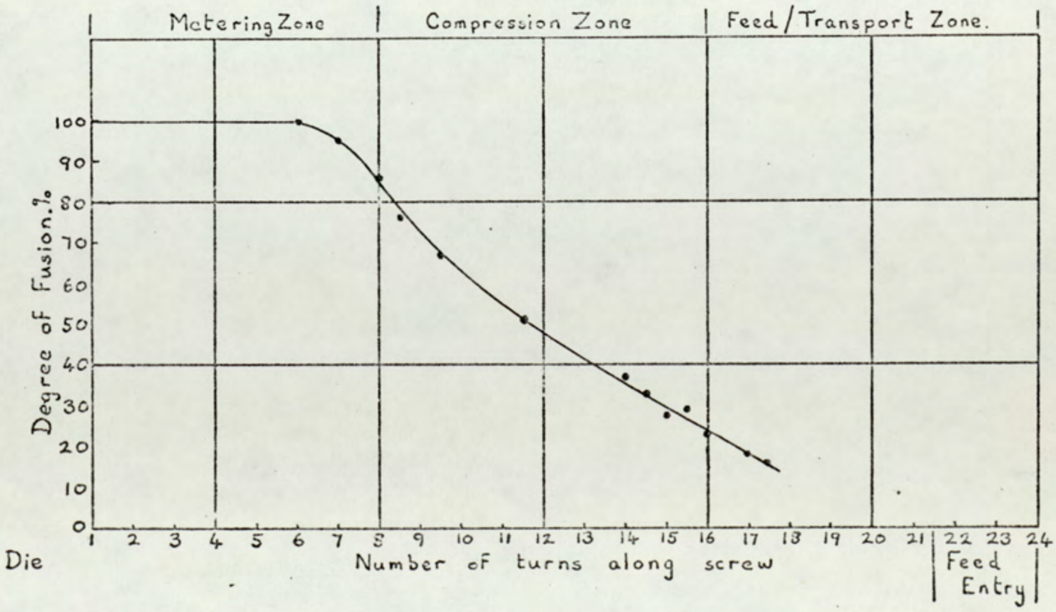


Fig. 53 Fusion rate - Lubricant: GMS 2.0/stearic acid nil. Screw speed 60 rpm

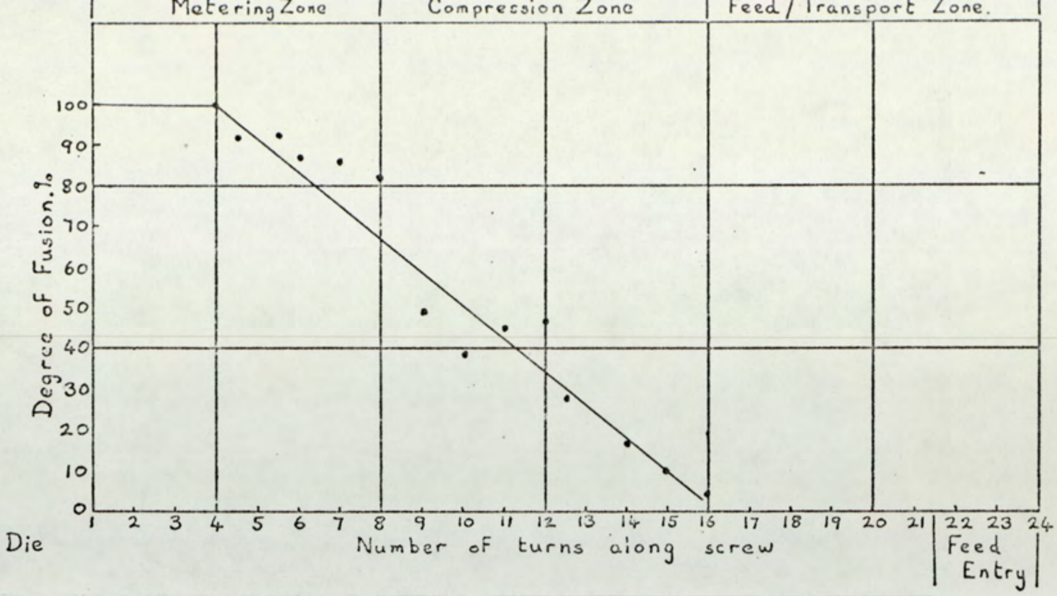


Fig. 54 Fusion rate - Lubricant: GMS 1.6 /stearic acid 0.4. Screw speed 30 rpm

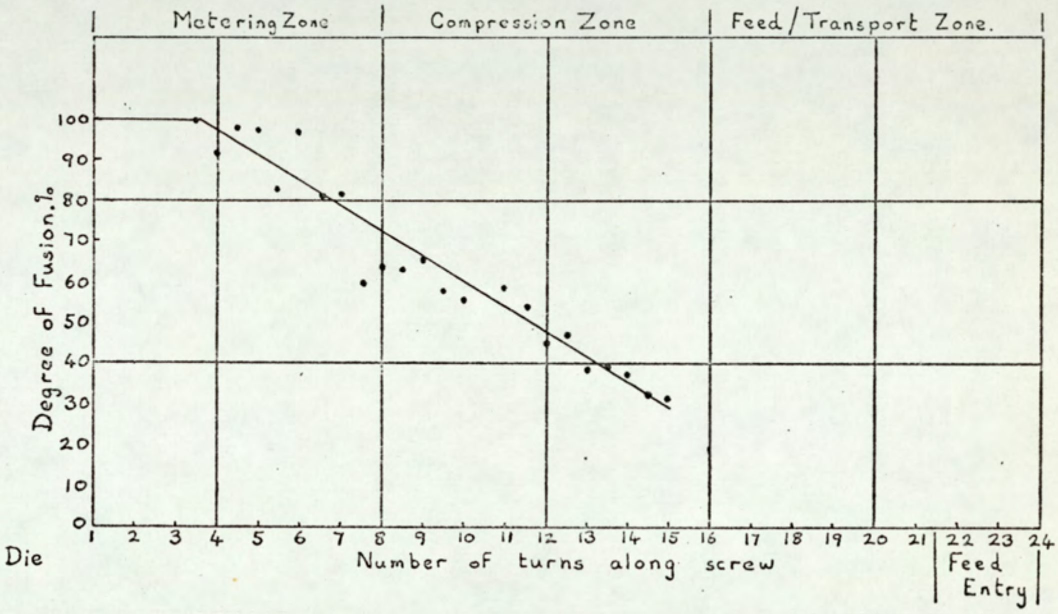


Fig. 55 Fusion rate - Lubricant : GMS 1.6 /stearic acid 0.4. Screw speed 50 rpm

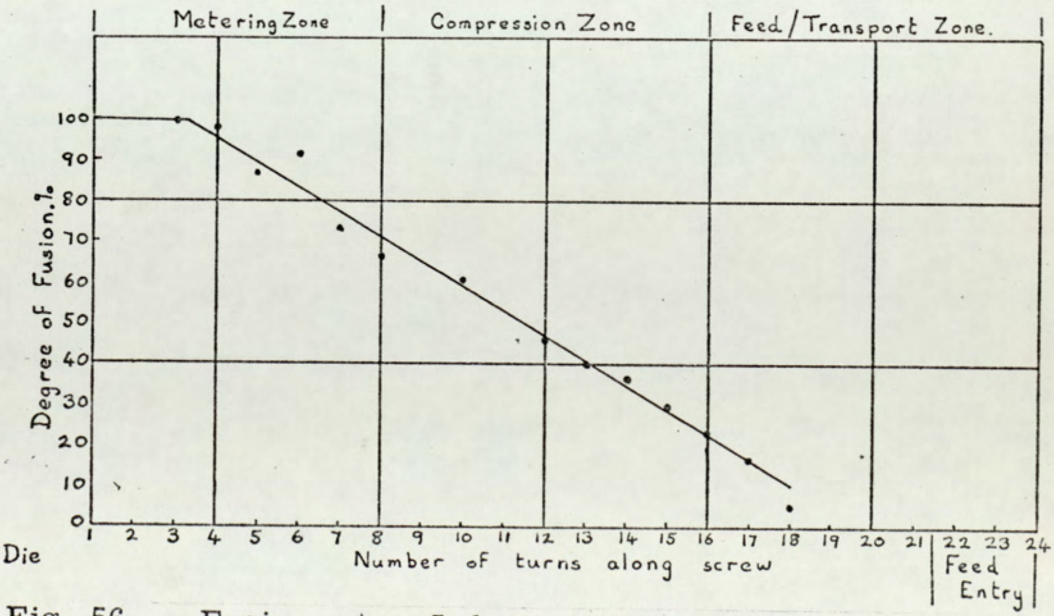


Fig. 56 Fusion rate - Lubricant: GMS 1.6 /stearic acid 0.4. Screw speed 60 rpm

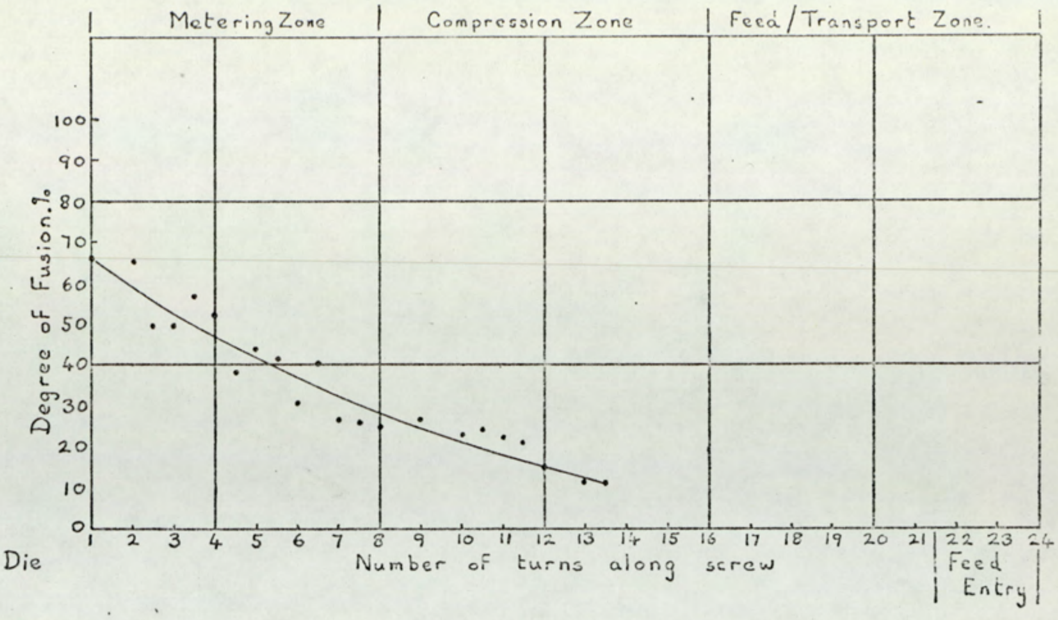


Fig. 57 Fusion rate - Lubricant: GMS 1.0/stearic acid 1.0. Screw speed 40 rpm

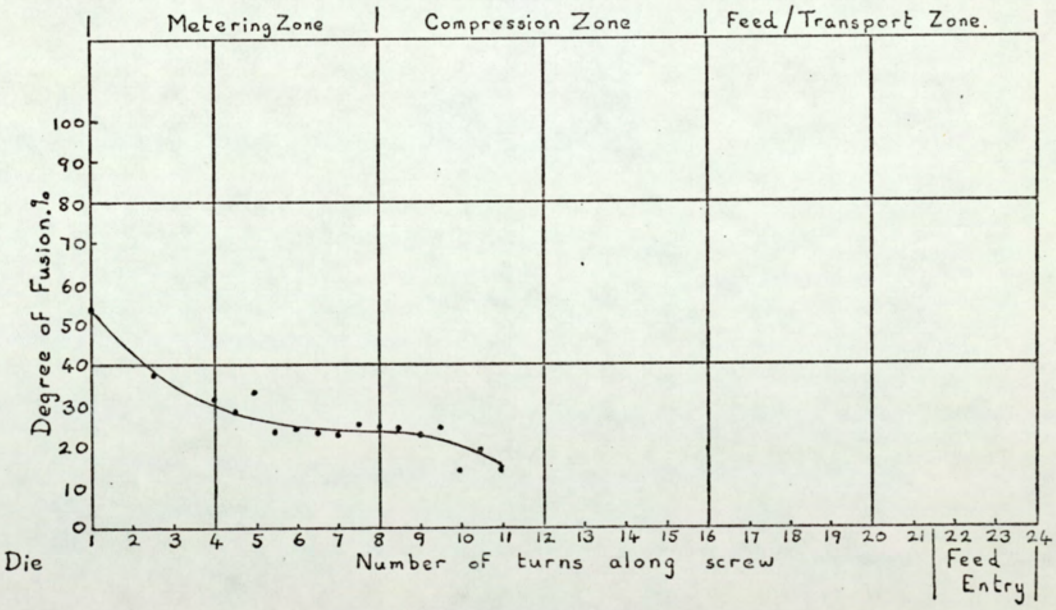


Fig. 58 Fusion rate - Lubricant GMS 1.0/stearic acid 1.0. Screw speed 70 rpm

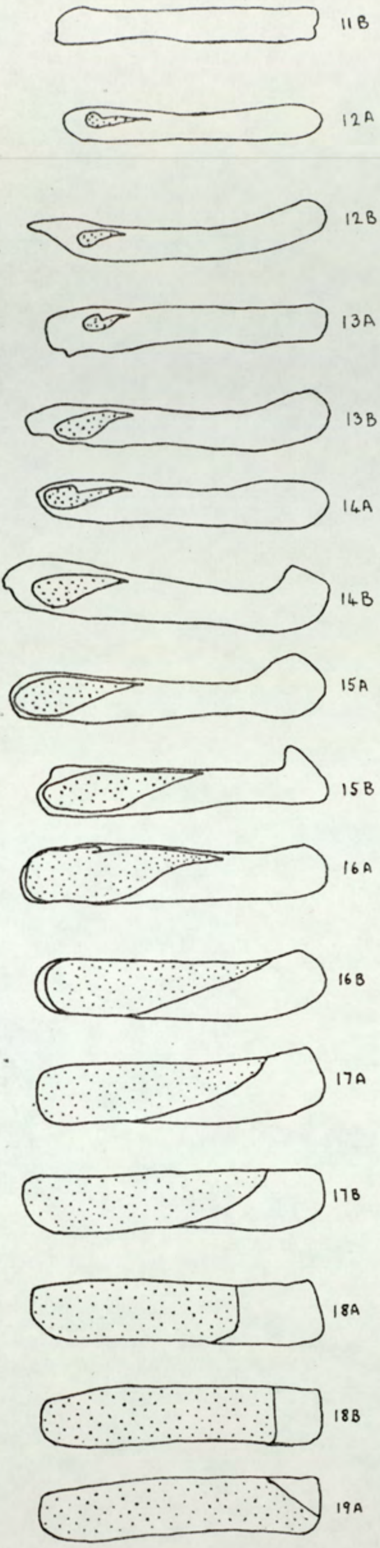


Fig. 59 Screw speed 20 rpm.

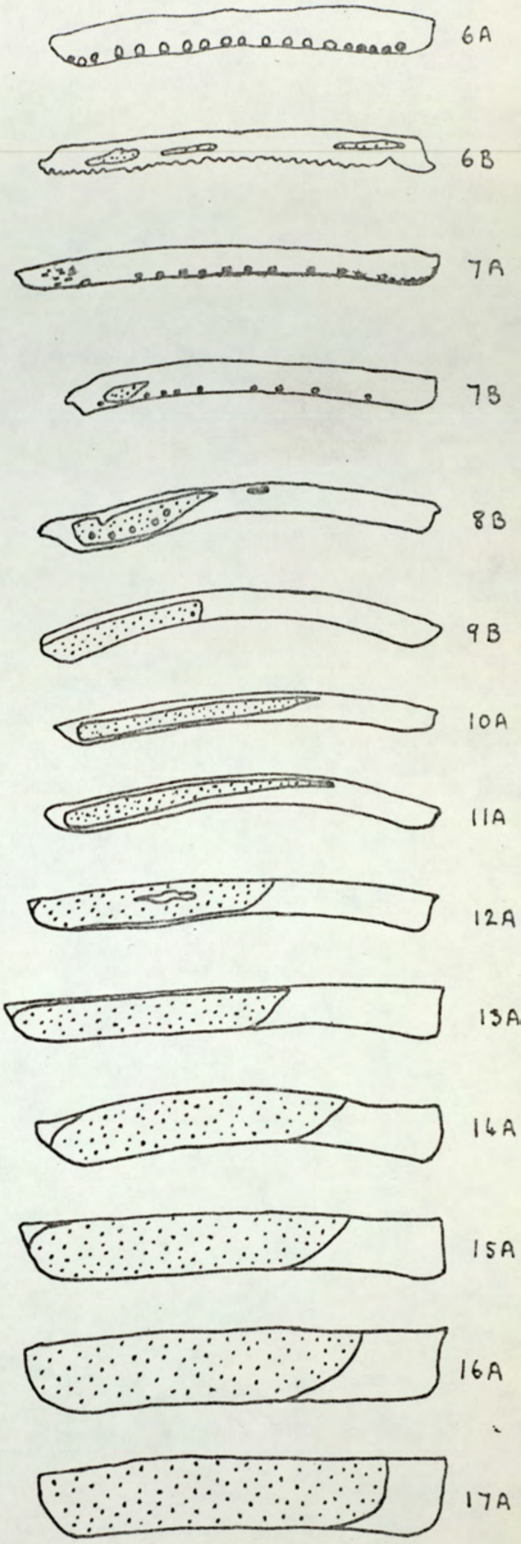


Fig. 60 Screw speed 60 rp

Screw channel cross sections - Lubricant: GMS 2.0/stearic acid nil.

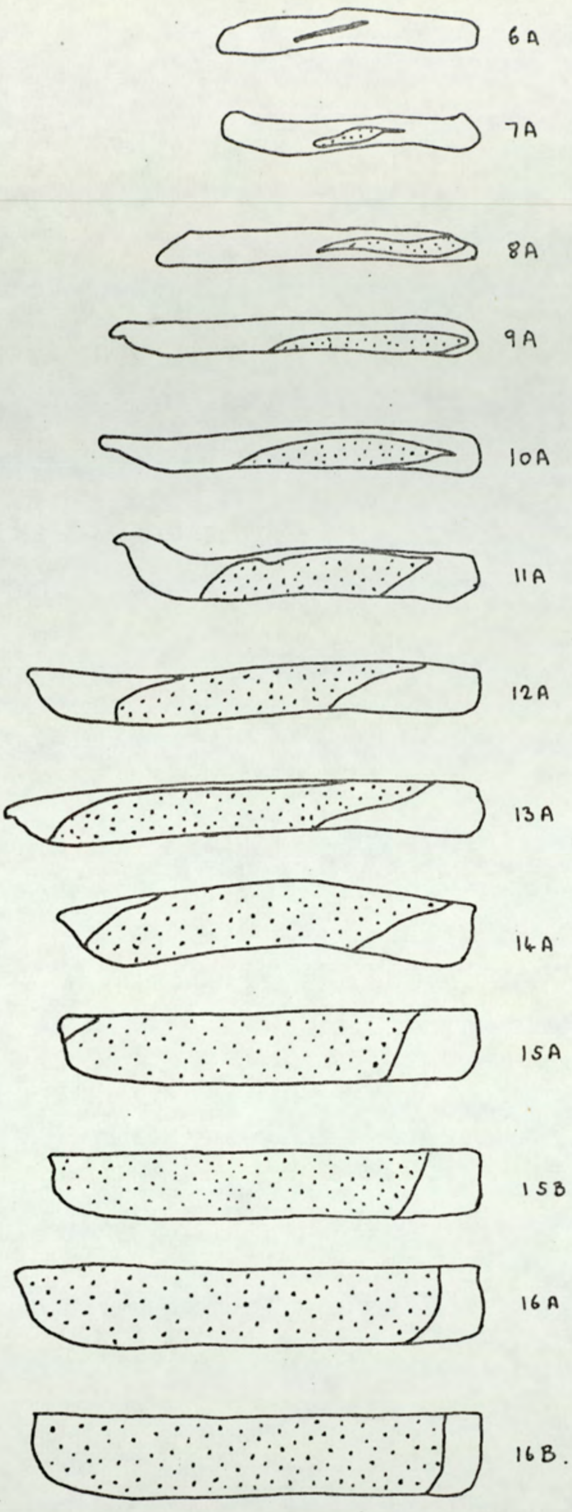


Fig. 61 Screw speed 20 rpm

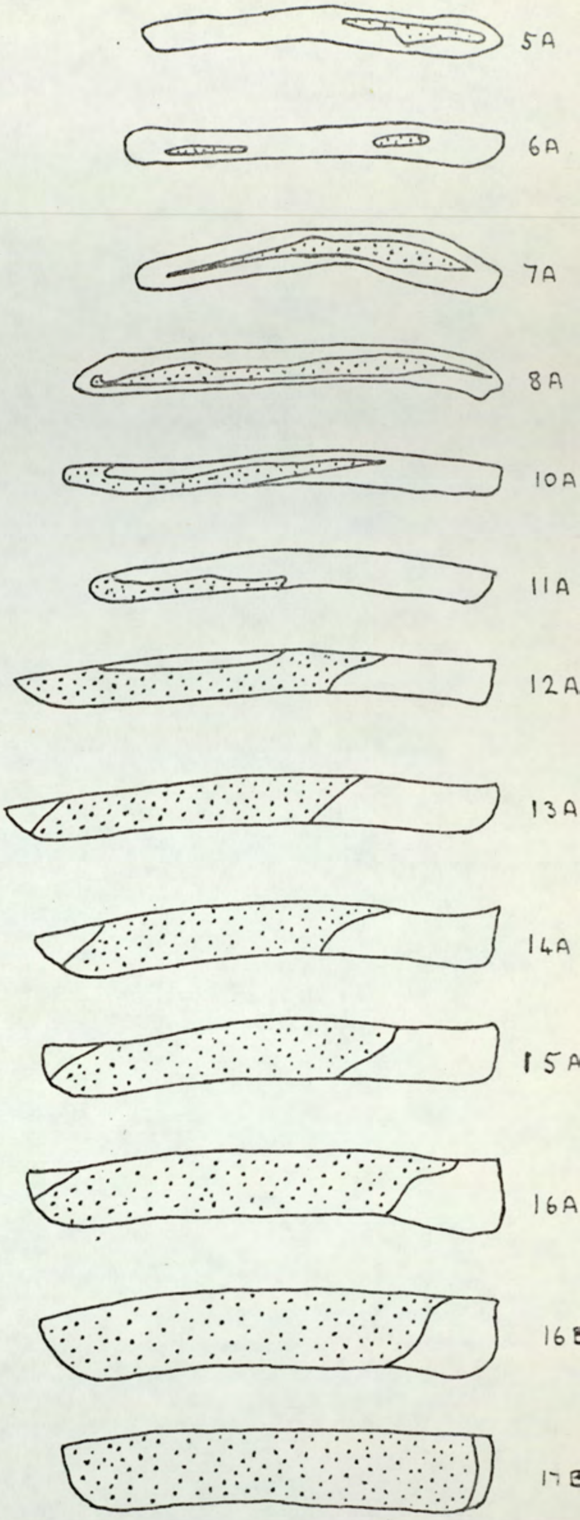


Fig. 62 Screw speed 60 rpm

Screw channel cross sections - Lubricant: GMS 1.6 /stearic acid 0.4.

It was found that at the higher screw speeds, air could be trapped in the melt, appearing in the early fusion stages as blisters and then as bubbles on the melt surface. Figs. 59 and 60 illustrate how increasing the screw speed results in the entrapment of air in the sintered powder. This air then becomes dispersed as small bubbles in the melt. Figures 61 and 62 are sections in which the part replacement of GMS by stearic acid has reduced the fusion rate. In this case there is no air entrapment but some forward fusion has occurred at the later stages, resulting in the spread of unfused material across the width of the channel.

5.7 Fusion mechanism

In the previous sections it has been shown how in the extrusion of PVC dry blends, the coating of PVC particles by the additives has influenced the rate at which conversion from powder to melt takes place as the material advances down the scew channel. It has also been shown that, depending on the compatibility of the lubricant, one or both of two fusion mechanisms can occur depending on the type and concentration of lubricant used. In order to understand the material requirements of a single screw extruder for PVC extrusion it

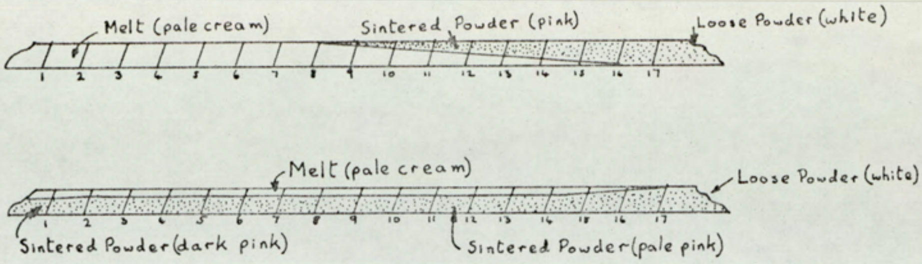


Fig. 63 Material unwound from screw channel showing rear and forward fusion mechanisms

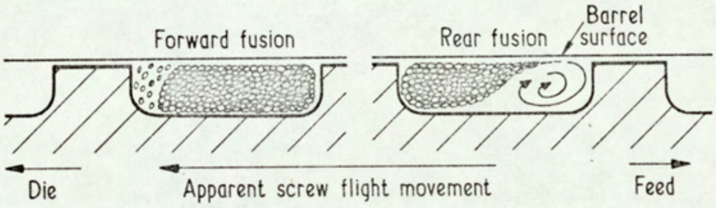


Fig. 64 Cross channel view of rear and forward fusion mechanisms.

appeared necessary to examine these two mechanisms more closely.

When the material was unwound from the extruder screw and viewed from the side formerly in contact with the screw core, the appearance of the material for the two mechanisms was as shown in figure 63 . In considering the position of the melt formation in relation to its position in the screw channel, then in the first case the melt collected at the rear of the channel, while in the second case it collected at the front of the channel. It was convenient to differentiate between them by describing the former as a "rear (of the channel) fusion model" (or mechanism), and the latter as a "forward fusion model".

The fusion mechanism was investigated by microscopic examination of successive sections taken at regular intervals from where fusion started to a point where nearly completed. Three procedures were adopted in which cross sections in line with the screw axis were cut and polished as described in the appendix The method mainly used was to store the polished sections overnight in stoppered glass specimen tubes filled with hydrogen sulphide gas to convert the lead stabiliser to lead sulphides. This being an intense black made outlines of unfused particles clearly visible at relatively low magnifications, while an even distribution of stabiliser (appearing as a fine grain) indicated a homogenous melt.

A second method was to extrude a PVC dryblend to which coloured granules had been premixed at a level of 5%. In this case the coloured granules were prepared by milling a proportion of the powder with a pigment on a two roll mill followed by removal as a 75 mm wide strip about 3 mm thick which were diced into cubes with a Girovinyle granulator. The third method was to add successive increments of different coloured dryblends into the hopper throat using weighed amounts of sufficient powder to fill two turns of the screw channel and covering a range of colours.

The polished sections were affixed to a glass coverslip using cedarwood oil (refractive index 1.51, which compares with 1.55 for PVC) which held the section in place by surface tension. This was then inverted and suitably mounted on the moving stage of a microscope for examination as shown in the Appendix.

5.8 Rear fusion model

For a number of experiments GMS was used alone as lubricant at a level of 2.0 phr with TBLS as stabiliser, and the rear fusion model occurred with rapid fusion rate as shown in Fig. 51. Examination of sections showed that the fusion behaviour was as follows.(see Figs. 63-65). The PVC powder mix was heated and compacted and became sintered together forming a solid mass. This travelled as a solid plug down the screw channel in accordance with the theory of Darnell and Mol, (63) and Tadmor (64). Fusion then occurred to material at the rear of the channel at the barrel surface and the melt was scraped off by the advancing flight to form a rolling mass at the rear of the channel. This effect is in agreement with the descriptions of Tadmor et al, Maddock, and Street, but an important difference was that melting was influenced far less by heat conduction from the wall and more by friction with the wall as shown by the effects of lubricants. The rolling melt pool then steadily grew in size at the expense of the solids bed until the channel was completely filled with melt. During this period it appeared that the melting differed from that absorbed by the authors previously mentioned in which granules melted as a result of heat conducted from the melt pool in addition to heat from the barrel. From the photomicrographs it can be seen that the boundary of the sintered powder is precisely defined with the appearance of semi-melted particles only at the boundary adjacent to the melt film against the barrel surface. Elongation of particles showed the areas where shear stresses were high, particles otherwise remaining approximately spherical. Particles remained spherical in the central areas of the sintered bed and at the surface of the bed in the vicinity of the screw surface, and at the forward edge. Some deformation occurred near the barrel surface, but the main area of shear was at the rear outer surface of the sintered bed. In this area the unfused PVC formed a tail of progressively elongated particles becoming thinner and thinner until a film was formed. This film eventually disappeared being transformed into a melt. Even during early stages of fusion when the sintered bed was relatively square in section a small tail and associated film formed near the barrel surface. The elongation of particles progressively increased as the tail became narrow as a result of the shear stresses progressively increasing with the convergence of the two melt streams. The effect was rather like passing the material through the tight nip of a two roll mill. The down channel section taken near to the base of the tail showed particles on the surface nearest to the screw surface to be approximately spherical, while those adjacent to the barrel appeared elongated similar to those in the cross channel section. The down channel section about half way along the tail showed particles near the barrel surface to again be very similar to the cross channel section, but the underside also showed elongation in a down channel direction.

This effect occurred both when the fusion had just started (as shown in photograph 2) where the "draw-down" effect is confined to a relatively small area of the sintered bed, and also at the last stages of fusion where only a small part of the sintered bed remained. In all these photographs (2-5) it can be seen that there is a thick film of melt enveloping the sintered powder. The existence of spherical particles over a high proportion of the periphery of the solid bed shows that any shear stresses applied to the particles must have been small, while the very clearly defined boundary between solid bed and melt shows that melting as a result of conducted heat did not occur. The surrounding melt film will have both insulated the solid bed from conducted heat, and probably of more importance, will have absorbed the shear energy applied by the rotating screw. The result would have been that the shear forces were lost as heat in the melt, giving unwanted rises in melt temperature which would have reduced its viscosity and therefore further reduced the possibility of transmitting shear forces to the sintered powder.

When successive colour changes were used the effect reported by Street (53) was found, where the solid bed moved by plug flow and advanced at a faster rate than the helically rolling melt. However, when uncoloured dryblends were followed by dryblends premixed with coloured granules, the relative positions of coloured granules in the solid bed and in the melt were usually no more than one diameter of screw length apart. The procedure was to run the extruder for various time periods to allow the granules to reach different points along the screw when stopped for screw ejection. The difference in results between the two methods was probably due to the fact that when successive coloured materials were added, the screw was incompletely filled with the result that compression of the loose powder to a compact plug increased its movement relative to the melt to produce an effect similar to that for granules in a short compression screw, as shown by Street.

A result of the overtaking of the melt by the solid bed was that materials of different colours were successively added to the melt from the solid bed so that the various colours in the channel cross section clearly showed the rolling path of the melt. The accompanying flow patterns from coloured material and the shear effects shown by elongated PVC particles (hydrogen sulphide treated as described above) in cross and down channel directions show that the melt moved in a helical rolling pattern in the rear of the channel. The melt also circulated to some extent in a cross channel helical path around the solid bed, particularly in the later stages of fusion.

The sintered powder moved as a solid plug with no cross channel movement other than in the tail as previously described. A slightly higher down channel velocity could occur at the centre as shown by the curvature of the boundary between colour changes. Coloured granules dispersed in the powder were deformed in a down channel direction only. This effect normally occurred following entry into the compression zone.

The overall mechanism for rear fusion is illustrated in Figure (65).

Forward Fusion

When dry blends containing more than a certain level of incompatible lubricant such as stearic acid or Wax E were extruded there was a different fusion mechanism. (The type and level of lubricant and extrusion conditions where this occurred have been described in section 4). In this case the fusion occurred by a mechanism described by Menges and Klenk (9, 10) where the melt pool formed forward of the solid bed. According to these authors this result was obtained for all of the large number of commercially available dryblends examined and even at extremes of temperature and shear conditions bordering on thermal decomposition, no more than 90% fusion could be obtained. It was considered that fusion occurred only to particles which had become wedged between flight land and barrel, and that the resulting melt collected at the forward side of the screw channel (Fig. 64).

In these investigations, examination of polished sections prepared as previously described, showed general agreement with the findings of Menges and Klenk although fusion of particles between flight land and barrel could not be conclusively proved. A zig-zag arrangement of unfused particles in the low pressure region just behind the screw flight land indicated a flow of material under pressure through this gap into the front of the channel. When the extrusion procedure of adding successive colours was used, the colour at the front of melt at the front of the channel corresponded to the colour of the powder on the opposite side of the flight. The coloured melt was spread in a down channel direction with no evidence of cross channel circulating flow so that a laminar effect occurred with a layer distributed adjacent to the solid bed of a colour corresponding to powder situated one or more turns ahead. In photograph 14 a strip of blue melt stretches back four turns behind the blue powder in the sintered bed. A layer of orange beginning at the screw tip is also similarly spread back over several turns. The elongation of green granules dispersed in the melt shown in photograph 15 indicates a considerable down channel distribution giving a velocity gradient effect with the slowest part near, (but not adjacent to) the solid bed. A possible overall mechanism is illustrated in figure 66

Assuming the screw is stationary and the barrel rotates, then the sintered powder moves as a solid plug in a down channel direction. As fusion proceeds it also has a very small velocity

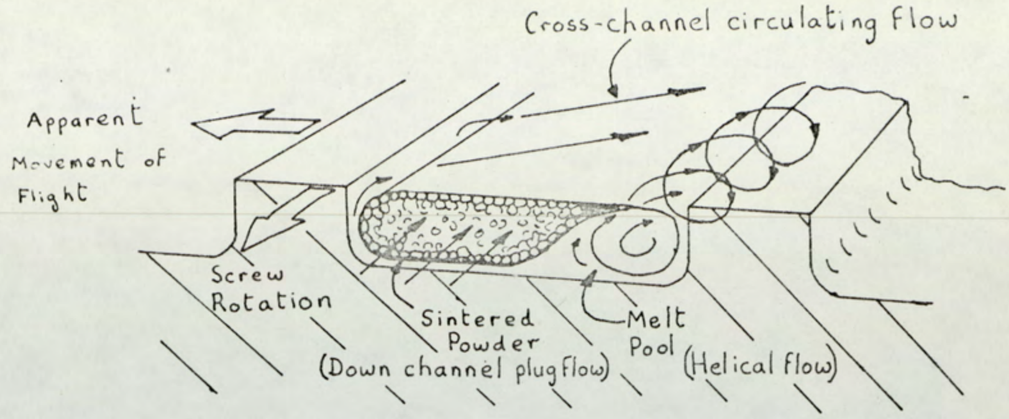


Fig. 65 Flow patterns during rear fusion

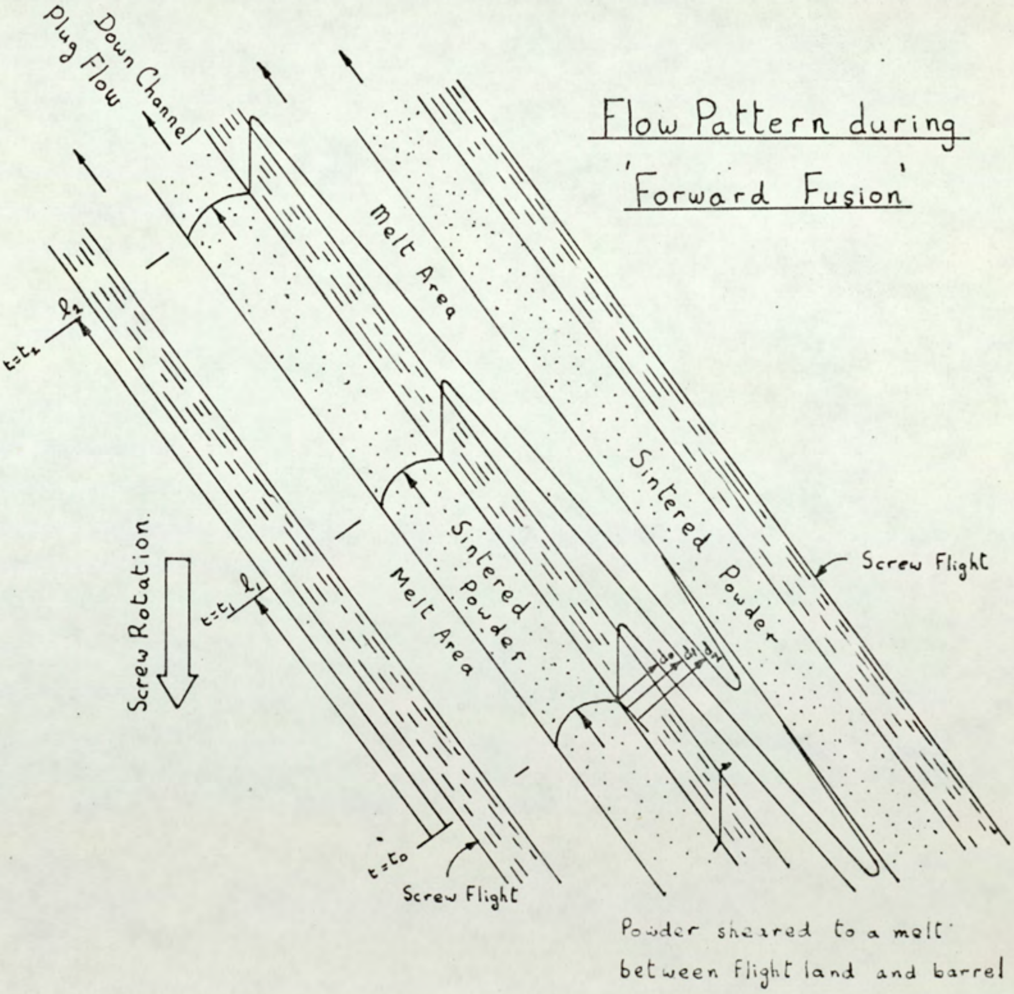


Fig. 66 Flow patterns during forward fusion

component in a cross channel direction, spilling over the flight as a melt into the forward area of the following channel. (The same channel, one turn previously, unless a multistart screw is used). The melt so formed travels relatively slowly with both a down channel component and a cross channel component which matches the solids bed cross channel velocity. If a colour change boundary moves in time t_1, a down channel distance l_1, then in this time a particle just melted will have travelled a distance d_1, as shown. During time t_2, corresponding movements will be l_2 and d_2 as shown. The boundary of the colour change in the melt will be situated after the time increments in the positions shown.

At the solid bed interface, drag from the plug flow will produce an increase in down channel velocity component of the melt. A similar effect is found with newly formed melt adjacent to the flight at the forward side of the channel. Cross channel velocity was generally greater at the barrel surface than at the channel base.

From photomicrographs it was further found that the melt contained a large quantity of individual "floating" unfused PVC particles. These particles were slightly enlongated and arranged in lines parallel to both the contour of the solid bed and to the down channel direction. The parallel strata shown in the photomicrograph of the channel cross section fills the full width of the melt pool showing that a rolling mixing action in the melt pool did not occur. It appears that cohesion between particles in the sintered bed was relatively poor and that they tended to break away and join the melt passing over the surface.

Photomicrograph No. 10 shows that a transition zone existed between the tightly packed solid bed and the melt whereas this was not so with the rear fusion model. The particles breaking away are more clearly shown in the down channel section. (No. 11).

With no rolling melt to provide the necessary shear forces for fusion of the sintered powder the fusion rate was relatively slow (as found by Menges and Klenk) and once a certain level of fusion was reached (usually less than 50%) the fusion rate was relatively small unless the influence of die back pressure caused some increases in fusion level. However, the values given in the graphs of percentage melt formed are optimistic as they include unfused particles dispersed in the melt.

As the forward fusion mechanism was incapable of providing complete fusion of particles to a melt, details of the mechanism and the values of fusion obtained at the screw tip were not of any real consequence compared with the rear fusion mechanism. The important data were the condition of formulation, and shear stress resulting from die and screw design, at which this mechanism occurred.

Effect of formulation, die and screw effects on fusion mechanism

The effect of lubricants could be determined by following the changes in fusion rate either as a compatible lubricant was replaced by an incompatible one or as the level of an incompatible lubricant such as Wax E was increased while used in combination with other lubricants in a typical commercial formulation.

Referring to Fig. 42, as the level of stearic acid was increased as incremental replacement of GMS, the fusion rate curve was displaced towards the screw tip, eventually not achieving full fusion. There was a transition between the relatively rapid fusion rate which gave complete fusion to a slow fusion rate which resulted in incomplete fusion. This transition in fusion rate was due to the transition in fusion mechanism from rear fusion model to forward fusion model. By referring to graphs 44, 47 and 48 and tables 10, 13, 14, the same effect can be seen for increasing levels of Wax E, DBLS and calcium stearate.

In the transition area, both models could occur simultaneously. Usually the rear fusion model started first, followed by the forward fusion model in the region of higher temperatures, while a return of rear fusion could occur as the screw tip was approached. The actual mechanism and combination of mechanisms were not usually precisely defined. During rear fusion, areas of unfused material could be dispersed in ragged patches on the surface formerly adjacent to the barrel becoming dispersed as unfused patches throughout the melt after the sintered bed had disappeared. On most of the occasions when this occurred, a sufficient screw length remained to fully fuse these unfused particles. In other cases it was found that as the rear fusion mechanism neared completion, the remaining sintered powder would break up into discrete areas of unfused particles floating in the melt as described by Tadmor et al (64) (Described by Stevens (65) as the iceberg region). On other occasions the solid bed would form a thin layer or several strata surrounded by melt. Examples are shown in Figs. 60 and 62 while patches of unfused particles can be seen in photograph 14 showing a cross section near completion of fusion by the rear fusion mechanism.

The scatter of the points in the graphs of fusion rates was in many cases a result of the modes of fusion. When fusion was a clear rear fusion model, then the points fell fairly closely on a straight line, but with forward fusion, a combination of mechanisms, or disintegration of a solid bed, the points on the graphs could be widely scattered.

The effect of additives other than lubricants appeared to depend on their compatibility and their cumulative effect with the lubricant, which could be additive or opposing. Generally, if the compatibility was relatively high the total lubricating efficiency was reduced, whereas additives of low compatibility had an additive lubricating effect.

In the case of conventional polymeric process aids / impact modifiers, the effect of the "rubbery" additive was to promote "adhesion" and therefore initiate a rear fusion mechanism and consequently give a relatively rapid fusion rate even when the lubricant system was such that it otherwise produced a forward fusion mechanism. Although the efficiency of the lubricant system in delaying fusion was opposed, the surface finish of the final extrudate was normally as good or further improved by the addition of such materials. A "less conventional" process aid appeared to have too low a compatibility and had an additive effect with the lubricant and produced a forward fusion model with associated low degree of fusion.

The overall effect of a precipitated calcium carbonate filler on fusion rate was similar to that of the process aid. In the original unfilled formulation, a forward fusion model occurred. When stearate coated precipiated chalk was incorporated, the rear fusion model occurred at all the levels examined (2.5 - 10 phr inclusive). In the case of the 2.5 phr level, fusion was barely completed at the screw tip as a result of the slow fusion of dispersed unfused patches, but with as little as 5 phr filler, fusion was rapid enough to give 100% melt at the entrance to the metering zone. As the total volume of addition in this case was comparatively small, its effect must be associated with its high surface area, but its exact function is ill defined. It will provide a large surface for the lubricant to cover, although its surface being calcium stearate will also be lubricating as experiments with calcium stearate showed in the previous section. At 5 phr the ratio of the surface area of the filler to that of the PVC polymer will (assuming particles in both cases to be spherical) be 75:100.

The effect of the stabiliser was not examined in this programme, but it was found in other work that this also depended on its compatibility and its influence on compatibility of lubricants in the formulation as a whole. For example, it was found that a liquid thio tin stabiliser, used for transparent products, promoted rear fusion such that a particular system producing forward fusion with a lead stabiliser produced rear fusion with the tin stabiliser.

The comparison of granules with powder emphasized the sensitivity of both the fusion mechanism and fusion rate of a powder feed to the lubricant coating on PVC particles. With Wax E used over the relatively wide range of 0.5-1.0 phr, the effect on fusion rate of granules was relatively small, as a consequence of the rear fusion model existing for the complete range. An additional point is that the fusion model resembled that described by Maddock and by Street and did not reproduce the mechanism of melting from a film formed from the sintered powder. With granules, a transition zone of solids to melt occurred with the result that a precise end point to the melting process could not be visually detected.

The effect of compression ratio was predictable. Increasing the screw compression ratio increased the shear stresses applied to the particles and therefore both increased the fusion rate for a given formulation and increased the level of incompatible lubricant at which the charge from rear fusion to forward fusion would occur.

There were no effects on fusion model due to the changes made in die land length.

5.11 Effect of fusion mechanism and rate on thermal stability

Although the exact chemistry of PVC degradation and stabilisation is in some dispute, the general method of stabilising low cost PVC formulations for opaque pipes and building products is to use lead stabilisers which behave as hydrogen chloride acceptors and therefore prevent the level of hydrogen chloride liberated reaching a concentration sufficient to catalyse a rapid degradation rate. In the degradation reaction, hydrogen chloride splits off by an unzipping reaction leaving conjugated double bonds in the PVC molecular chain. If the number of consequtive conjugated double bonds is greater than seven then the PVC appears pink in colour. The release of 0.1% hydrogen chloride will produce this effect. The thermal degradation of PVC is both time and temperature dependent so that the higher the temperature for a given time and the longer the time the PVC is held at a given temperature the greater will be the degree of degradation.

In stabilising PVC dryblends with solid lead stabiliser, the stabiliser will remain as a coating of solid particles on the surface of the PVC particles until dispersed throughout the PVC when the polymer is fused into a homogenous melt. This factor was exploited, as described previously, to identify unfused PVC. This means that the longer the solid bed is held at an elevated temperature before melting then the more advanced will the degradation become during extrusion. Thus when a rear fusion mechanism occurred giving a rapid fusion rate, the stabiliser was quickly incorporated into the PVC and able to function effectively, but when a forward fusion mechanism occurred the stability depended on the diffusion of hydrogen chloride from inside the PVC particles to the lead stabiliser at the particle surface. For both mechanisms the PVC powder in the sintered state became pink in colour as it was advanced down the screw channel. Following fusion, it was transformed to a pale cream melt, this colour being a result of the pigmenting effect of the dispersed lead stabiliser. If the rear fusion was very rapid there was a tendency for the melt to have a torn appearance due to drag at the barrel surface and the conversion of mechanical energy into shear heat resulted in elevated melt temperatures which produced a deep cream - yellow melt, eventually becoming pink brown which frequently resulted in material stagnating and degrading completely in the die. Higher screw speeds also produced this effect.

When the forward fusion model occurred, the sintered powder being conveyed the full length of the screw became a deep pink colour, and imparted this colour to the extrudate. A further effect was that any melt formed by a forward fusion mechanism, either by forward fusion alone or in combination with a rear fusion model, contained floating unfused PVC particles which were in an unstabilised condition.

The optimum fusion for good thermal stability was a rear fusion model which produced complete fusion but with sufficient lubrication to prevent too rapid a fusion and so avoid excessive melt temperatures.

6. Discussion

The purpose of this section is to bring together the results which have already been described in detail in the respective sections above and discuss their implication regarding the operation of single screw extruders with rigid PVC dry blends. Comparisons are also made with published data summarised in the review comprising section 3.

The objectives were to consider which of the many possible variables were the most decisive in determining the success or failure of PVC powder extrusion and to gain a better understanding of the mechanisms involved so that the particular variables could be better controlled. Two types of variable exist. The first type are the preselected production conditions of screw and die design, formulation, screw speed, extruder temperature, and powder mixing conditions. The second type are the "uncontrollable" variables which can occur with temperature, screw speed and raw materials.

The results of this investigation show that the problems of dry blend extrusion can be concentrated around the properties of the feed material which influence its conversion from a powder to a melt. The basic problem in using PVC powder, as shown by the results in section 5, is that the presence of lubricants on the surface of the PVC polymer particles, (following the powder mixing process) controls the level of particle surface friction, thereby determining the melting effect. Heat and pressure alone produced a sintered material with very poor physical properties. For a fully homogenous melt, a shear stress sufficient to break down the particles needs to be applied, and this stress cannot be reached if the lubrication is too efficient.

6.1 Fusion model

Menges and Klenk (9, 10) concluded that the basic differences between extrusion of PVC dryblends and other common thermoplastics such

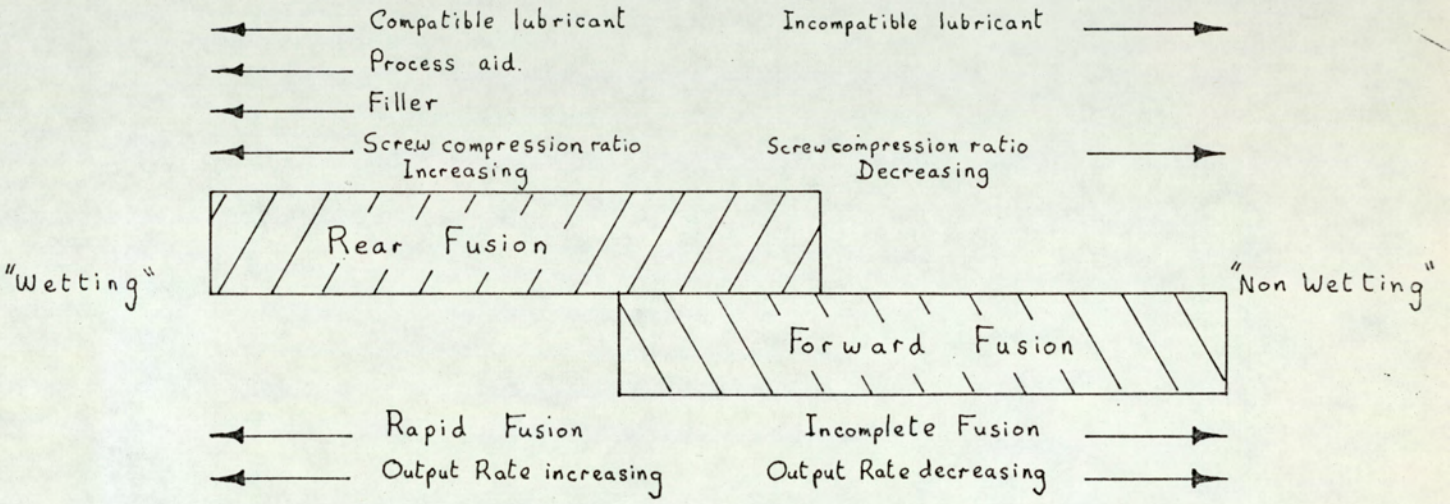


Fig. 67 Summary of fusion effects

as polyethylene was that the former was "non-wetting" (or non-adhering) and latter were "wetting" (or adhering) materials. By this they meant that PVC would not adhere to the metal surfaces but would slide as a solid plug along the length of the screw channel, melting occurring only to the proportion of the material trapped between flight land and barrel surface. This proportion varied between 10 and 80% so that at least 20% of the material remained unfused. In conflict with their findings we have literature references to single screw extruders running satisfactorily on dryblends, although mainly in the United States.

This anomaly is explained in terms of the effects of composition variables reported above on the melting model. If the compound produced the conventional rear fusion model of "adhering" polymers then fusion was rapid and normally complete. If the compound was "non-wetting" with the resultant forward fusion model of Menges and Klenk, then melting was invariably incomplete. The formulation effects were unfortunately not so precise as intermediate stages could occur. Even when the required rear fusion model was obtained the fusion rate was still important for considerations of output and thermal stability. The influence of the screw design further reduced the limits of operation.

Considering firstly the more widely used formulations which exclude process aids and fillers either on cost or performance requirements, then providing the level of incompatible lubricant is below a certain level, (preferably in combination with a compatible lubricant) the rear fusion model will occur and the melt will be homogenous. As soon as the level of adhesion is reduced, either by increasing the total level of incompatible lubricant or by using a lubricant of lower compatibility then the rear fusion model will no longer be obtained. Jones and Hawkes (14) statement that the function of lubricants as gelation controller is more important than the prevention of material sticking in the die during twin screw extrusion applies probably even more critically to single screw extrusion.

6.2 Formulation effects

In Menges and Klenks experiments (9, 10) the similar unsatisfactory results for a number of commercially produced dryblends indicates that they were formulated as a result of satisfactory performances on twin screw extruders and were all totally unsuitable for the single screw machine. This effect also throws doubts on the conclusion reached from results showing creep properties of twin screw extruded pipe to be superior to that produced on the single screw machine using the same tube die and dry blend (68).

However, in terms of changes in levels of particular lubricants the effect on physical properties for twin screw extruders appears to be similar to those for single screw extruders. A satisfactory product can be rendered unsatisfactory by a relatively small increase in the level of incompatible lubricant. The relatively large changes in impact strength with very small changes in dibasic/normal lead stearate ratio shown by Jones and Hawkes show a similar sensitivity to the large effects on fusion rate with small lubricant changes in the single screw extruder. These two effects are probably related.

When a single lubricant which was compatible (e.g. GMS) was used at a level of 1.0 phr, then both the surface of the melt extracted from the screw channel and the extrudate surface were rough. If the lubricant was incompatible (e.g. stearic acid) then the melt and extrudate surfaces were both smooth.

However, when GMS was used at levels of 2.0 and 2.5 phr the melt surface and extrudate surface were now smooth, but the fusion rate was comparable with that at the lower levels of GMS. Various explanations can be considered. The PVC may be plasticised by the GMS and flow more easily. The surface of the PVC particles may become sticky. The PVC surface may slip but still have sufficient affinity to metal surfaces to provide forces sufficient for particle fusion. From the photomicrographs it appeared that in the rear fusion model, interparticular adhesion is important for formation of a melt film from the solid bed, but for this to occur, forces need to be transmitted to the sintered powder from the barrel and screw surfaces. This area needs further investigation to establish the lubricant function more accurately.

Stearate coated precipitated chalks are now widely used at levels up to 10 phr to provide improvements in impact strength and ease of processing at no extra cost. However, DIN (German) standards prohibit their use in water pipe presumably due to adverse stress rupture properties

properties while for sewer pipes a limit is set at 3%. Results by Jones and Hawkes (14) showed improvements in impact strength of twin screw extruded tubes with increasing filler level, passing through a maximum at 7.5 phr. The increase in power consumption with increasing filler level could be due to the faster fusion rate shown by the Brabender results of the same authors. The results in section 5 show that the same type of effect occurred with the single screw extruder. As 2.5 phr was a sufficient filler level to change the fusion model from forward to rear, this relatively small addition was able to bring about a considerable increase in fusion rate, while the relatively rapid fusion rate brought about by 5 phr and 7.5 phr filler levels appear to ensure complete fusion. The reduced output rate of the single screw extruder will be due to the earlier completion of fusion. Although the results of Jones and Hawkes for the twin screw extruder also show a reduction in output rate with increasing filler level, the same considerations concerning the relative contributions of plug flow and cross channel flow effects will not apply. The output rate of the twin screw extruder should be directly proportional to bulk density of the feed material.

The exact function of the fillers in terms of either modifying either the lubricant efficiency or wetting effects is not clear, although the possible increase in surface area for lubricants to cover as suggested above seems a reasonable explanation. If the stearate coating was alone responsible for filler effects then the opposite behaviour with regard to fusion rate as shown for calcium stearate would apply.

The effects of polymeric process aids agreed with Jones and Hawkes'

Brabender Plastograph results (14), in that for both cases they were shown to promote earlier fusion. The existence of polymeric additives as a separate phase could well increase wetting as they normally contain an elastomeric component. However they normally improved the surface gloss, whereas elastomeric blends used as toughening agents in polystyrene and ABS have the opposite effect. Although, from the literature, problems of dispersion of these additives can occur, there was no evidence of this from the microscopic examinations, but this was not specifically investigated.

6.3 Effects of "uncontrollable" variables

The above results show that output rate depends on fusion rate, the later the attainment of complete fusion, the higher the output rate. Small variations in any part of the process which affect the shear stress on the material, in particular the lubricant level, will cause fluctuations in fusion rate with consequent changes in output rate. In the case of pressure pipe this could cause high scrap rates. If the incompatible lubricant level is at a maximum consistent with a retention of the rear fusion model, then the slightest increase in lubricant level or incompatibility (batch to batch variation) will result in incomplete fusion of the PVC.

Two literature references comment on the disadvantage of single screw extruders due to fluctuating output rates as a result of temperature variations. This also could well be due directly to variations in fusion rate. Limitations of output rate appear to be due as much to air entrapment as it does to development of excessive shear heat in the melt at the higher screw speeds. However, screw speed appeared to have far less influence on fusion rate than the other variables. Comparisons of powder with granule extrusion show that the latter method is far less susceptable to formulation or temperature variables. In addition variables resulting from particle flow behaviour will be eliminated. Air inclusion is also far less likely to occur.

6.4 Heat stability

Most opaque rigid PVC building products use lead stabilised formulations. The lead stabiliser consists of solid non-melting particles in the 1-5 μ range. Up to the point where fusion occurred, the stabiliser existed as a powder coating on loose or sintered PVC particles of 150 μ diamater. This meant that during the sintered powder stage, any hydrogen chloride liberated had to travel distances up to 75 μ through the PVC before reaching the stabiliser. The stability at this stage must in some way be dependent upon the internal structure of PVC particles which have varying degrees of porosity. Examination of the material removed from the channel of the extracted screw showed that the sintered powder progressing down the channel changed from white to a deep pink colour, a feature used for microscopic identification of areas of unfused material.

Once fusion had occurred, the pigmenting action of the dispersed lead stabiliser coloured the melt while or pale cream. The result was that when the rear fusion occurred, rapid incorporation of the stabiliser produced a well stabilised melt. Where very rapid melting occurred, the melt surface appeared rough where it had dragged against the barrel surface. In addition, the extrudate was deep yellow/pink, often with a rough surface and the melt tended to stick and decompose in the die.

When the pink area of unfused sintered powder was examined under the microscope, the particles were found to have pink centres, i. e. degradation was occurring in the central areas of the particles due to their remoteness from the peripheral stabiliser network. This network appeared to prevent "castrophic" degradation.

It is evident that the optimum fusion rate concerning homogenity and output rate is equally important when considering heat stability. When considering that so many rigid PVC extrusions are used for building products then processing factors will be important if they influence long term weathering properties in addition to immediate physical properties and appearance. Both the depletion of stabiliser with rapidly fusing compounds and poor distribution of stabiliser in underfused products could well have adverse effects, as also could degradation during processing from overheating or poor stabiliser distribution. The presence of conjugated double bonds as a result of processing degradation will provide weak sites for degradation to continue under the influence of ultraviolet light and atmospheric oxygen.

6. 5 Comparison of powders with granules

Advantages claimed for extrusion of powders in place of granules include better homogenity, improved product properties, lower processing temperatures, and cost savings from reduced levels of stabilisers and lubricants (12). Products were said to be more uniform as the varying degree of granule fusion was removed. The heating rate of powder was also said to be superior to granules (28).

In these experiments there was no evidence that the heating rate of powder was superior to granules. In fact the heating rate of the bulk of the material depended on internal friction from melted material, and this was dependent on fusion rate. A consequence of the effects of variables on fusion is that powder extrusion is likely to have poorer homogenity and less uniform, products than granules. In view of the need with powders to convert a blend of particles to a melt rather than simply transform a premelted material to a new shape as in the case of granules, it is difficult to see how processing temperatures can be lower for powders. However, with a single pass through the extruder, lower stabiliser levels may be possible provided stability during subsequent weathering is adequate. Lubricant levels could in some cases be lower but this will depend on control of fusion rate.

The fusion rate results in section 5 showed that for granule extrusion, this was comparatively insensitive to lubricant level when compared with powder extrusion. The limits of formulation variables and extrusion conditions are therefore relatively wide for granule extrusion and the process a more reliable one.

With the rather narrow limits of optimum fusion rate for which an extrudate could be obtained, there was no evidence of rippling due to overgelation (14). As very rapidly fusing materials produced an unacceptably rough or degraded extrudate it is possible that the rippling phenomenum is confined to twin screw extruders. Rippling did sometimes occur with forward fusion, this being a result of differing swelling behaviour between the separate areas of fused and unfused material.

6.6 Dryblend mixing variables

Production high speed mixers normally have one fixed high speed and control is by final mix temperature. At this point the mix is discharged to a cooler. The only condition that can therefore be usefully varied to obtain better products or better economies is the discharge temperature. In general the results of effects of discharge temperatures on bulk density agreed with published results i.e. bulk density increased with increasing mixer discharge temperature. However, the high bulk densities obtained by Morohashi (43) by using discharge temperatures of 175°C were not attempted as degradation occurred at the high temperatures. Morohashi (43) claimed that good results from powder feeds on single screw extruders were possible providing high bulk densities obtainable from 175°C discharge temperatures were used. Jones and Hawkes (14) showed how increased bulk density increased output rate and impact properties when using a twin screw extruder. In section 4 above, results for the single screw extruder varied, probably as a result of three different effects. The general effect was that increased bulk density resulting from increased mixer discharge temperature produced an increase in output rate, but where powder flow properties were adversely effected by a sticky stage, this effect was superimposed. A further effect which occurred with the denser polymer was a fall in output for mix temperatures above 80°C which was probably due to its earlier fusion point, i.e. the same effect at that shown for varying levels of lubricant, in a later section. Overall, it appeared that with single screw extruders, very large increases in discharge temperature are necessary to produce any really significant effects on output rate, while fusion rates are also influenced very little for a range of discharge temperatures above 100°C.

Jones and Hawkes (14) recommended that the polymer requirement varied between different types of twin screw extruder. As single screw extruders are normally flood fed (in contrast to twin screw machines which usually have starved metered feeds) advantage can be made of the higher bulk density polymers with their better flow properties while their ruggedness make choice of polymer generally less critical.

Recent developments are aimed at extruding either from "warm mixed" powder, using temperatures of about 80°C, i.e. just above the melting point of the lubricating waxes, or from powder pre-mixed at room temperature. The results of the effects of mixer discharge temperature on flow properties show that with certain lubricants, the warm mixed materials use the worst possible discharge temperature for producing good powder flow properties.

The reported drop in motor power of the high speed mixer after several minutes mixing was observed and found to coincide quite well with the lubricant melting point and was considered not to be due to the reported effect of swelling of the PVC (43). Previous work using pigmented lubricants substantiates this conclusion (68).

6.7 Comparisons of single and twin screw extruders

Results of the factory surveys in section 2 showed how single screw extruders were overall as good or better than twin screw extruders during a period when all single screw extruders and the majority of twin screw machines were granule fed. The effect of formulation variables on mode of fusion and the variability of output rate resulting if fusion rates vary, are probably the major reasons for the decline in the use of single screw machines with the introduction of direct powder extrusion of rigid PVC in Europe. The comparative success of single screw extruders in the USA could well be due to the combination of legislation prohibiting lead stabilisers in favour of organo-tin types and the development of two stage screws and vented barrels. The tin stabilisers will promote "wetting" and therefore rear fusion while the two stage screw can "even out" small output rate fluctuations resulting from fusion rate variations. Venting will eliminate air inclusion problems. There is also in the USA a relatively large requirement for rigid PVC packaging films, in which modifiers are normally used. As these materials were shown above to promote rear fusion, single screw machines would be expected to be satisfactory for this application as well.

It is probably for similar reasons that single screws are used successfully for injection moulding and bottle blowing. For reasons of either flow properties or clarity requirements, process aids and compatible lubricants are normally used. Variability in output will be less critical as injection moulding involves intermittent screw operation, while output rates in blow moulding are frequently less critical than in other extrusion processes.

Dahlmann commented that single screw extruders were satisfactory for thin wall tubes as high die pressures are produced (31). This effect would be to "even out fluctuations in screw output as well as to produce high shear stresses for acceleration of fusion rate.

Although the critical nature of the lubrication effects appears to make the single screw extruder rather unfavourable when compared with the twin screw extruder, it is possible that the twin screw extruder is not so much better in this respect. It is likely that the twin screw extruder is able to produce inferior but commercially acceptable products because any unfused material would be distributed to an extent sufficient to achieve a visually acceptable product with physical properties only just meeting a specification (where one exists). The progressive reduction in impact results of Jones and Hawkes (14) with increasing proportion of lead stearate suggests that this is the case.

6.8 Uses and Limitations of single screw extruders

This, to some extent, is explained in the comparison of United States and European practice, resulting from the different wetting characteristics of the compounds used on either side of the Atlantic. The single screw extruder appears to be satisfactory for granule extrusion but, for powder extrusion, limitations apply in choice of fomulation, variability in output rate, air inclusion and development of excessive shear heat. With precise control of formulation, venting and improved temperature control (including cooling systems,) there appears to be no reason why single screw extruders such not be used economically for direct powder extrusion. Whether a single screw extruder equipped to all these requirements would be a more economic proposition than the twin screw type would need to be determined.

7. Conclusions

7.1 In changing from a granule to a powder feed for a single screw extruder, the formulation limits become very narrow, mainly as a result of the influence of the additives on rate of fusion of powder to a melt.

7.2 The additives having the greatest influence on fusion rate are the lubricants, although all the additives must be considered including stabilisers, fillers, process aids and impact modifiers.

7.3 Depending on the additives used, one of two fusion models can occur. In the rear of the channel fusion (which is that normally obtained for polyethylene and plasticised PVC), fusion is relatively rapid. In the second case material fuses only when trapped between flight land and barrel and the melt collects at the forward side of the screw channel. This mechanism is insufficient to obtain complete fusion of the PVC.

7.4 Incorporation of compatible lubricants, filler, process aid or impact modifier promotes rear fusion, while incompatible lubricants promote forward fusion. A balance of incompatible lubricant to the rear fusion promoting additives is necessary to provide a fusion rate fast enough to achieve a homogeneous melt but not so fast that overheating and degradation occur.

7.5 Variability in fusion rate will produce variations in output rate. Strict control of all variables, particularly lubricant level is therefore necessary to achieve good product dimension control.

7.6 The sensitivity of single screw extruder processing behaviour to the effects of additives explains why twin screw extruders became more acceptable than single screw machines when (for economic reasons) changes from granule to powder feed were made. The results also explain why the different requirements for rigid PVC products has resulted in the continued use of single screw extruders for powder extrusion in the U.S.A. whereas in Europe twin screw machines are normally used.

7.7 Effects of high speed mixer discharge temperature showed that attention must be paid to such effects on flow properties. Recent developments have been towards extrusion of "warm mixed" blends in which lower discharge temperatures are used there by increasing plant capacity. However this practice could produce blends having the minimum rate of powder flow in the hopper with associated problems of "bridging" and "rat-holing".

7.8 Polymer degradation sufficient to produce pink colouration was found to occur as a result of the solid lead stabilisers being situated as a surface coating on the PVC particles for a substantial distance along the screw channel. Degradation will be at a minimum for an optimum fusion rate whereby incorporation of lead stabiliser is sufficiently rapid to efficiently stabilise the material, but not so rapid that excessive temperatures are generated within the melt.

7.9 Although the mechanism of compatible lubricant, process aid, impact modifier and filler on promoting rear fusion is not clear, the qualitative effects that have been determined are very important aspects for understanding single screw extruder problems. By using the laboratory scale extruder techniques described, any new additive can be assessed in comparison with existing materials.

8. Future work

The influence of lubricants on the reported "wetting" and non-"wetting" effects and the intrinsic lubricant properties require a more quantitative measurement. This would make elimination of unsatisfactory materials from the very wide range offered much easier, while "tailor made" lubricants would be possible. This should reduce the large amount of "trial and error" development necessary at the moment. Among laboratory techniques new to this field is the annular shear cell, which can be used for measuring friction properties, simulating extruder conditions of hot metal surfaces and shear rate effects.

9. Glossary of terms

9.1 Dryblend "A free flowing blend of PVC powder and additives produced under the action of heat, but without the creation of pressure". This definition is taken from VD1 Berichte 130-1968 (1) A high speed mixer is normally used with mix temperature reaching 100-150°C.

9.2 Pre-mix or Cold blend - This term is used when the blend is mixed without any appreciable rise in temperature. Either high speed or low speed mixers can be used.

9.3 Warm blend - This is similar to a dry blend except that the mix temperature is restricted to an intermediate range of 50-80°C being sufficient to melt certain lubricants.

9.4 Fusion - This term is used to describe the conversion of secondary PVC particles to a homogenuous mass.

PVC differs from most of the common polymers in that the polymer is insoluble in the monomer. The result is that polymer is precipitated forming 5μ particles within the suspension droplet which coalesce together to a certain degree depending on polymerisation conditions. The suspension droplet forms into a particle of about 150μ (termed the secondary particle) having a skin containing residues from suspension agents while the central regions contain the coalesced 5μ particles (termed primary particles). The primary particles are spherical, while the secondary particles can vary from "approximately spherical" to quite knobbly (resembling a potato). The primary particles appear to melt at about 190°C without the application of a shear stress (2). The term "melting" has not been used to describe the conversion of PVC secondary particles to a molten mass as this infers that heat alone is sufficient as is the case with wax, polyethylene, and polystyrene, whereas these PVC particles require the application of a shear stress in addition to heat.

The terms "gelled" and "plasticised" are also sometimes used but should be confined to processes involving plasticiser absorption. The term "plastified" is an American one which probably is the best to describe this situation but so far has not had wide acceptance in Europe.

"Fusion" does not appear to be a clearly defined term and could well be the result of the translation of German terms describing work in this field using the Brabender Plastograph (4). However, as this term is so widely used at present is has been adopted for this work.

9.5 Plasticised PVC - This describes PVC to which a plasticiser has been incorporated to make it flexible, usually between 25% - 50% of the total weight.

9.6 Unplasticised PVC (or PVC) - PVC without a plasticiser.

9.7 Rigid PVC - This describes PVC by contrasting it with flexible PVC which was widely used for many years before rigid PVC came into use. With earlier processing machinery and for some products such as thin clear sheets small amounts of plasticiser are used so that although the PVC is still rigid (it can even be very brittle) it is no longer "unplasticised".

9.8 Stabiliser

An additive to curb thermal degradation. Ideally it should prevent degradation but it appears that some inevitably occurs.

9.9 Lubricant

An additive which reduces adhesion of the PVC to any internal machine surfaces, chiefly, screw, barrel and die.

9.10 Process aid

An additive (usually polymeric) which produces a smooth glossy extrudate which is easier to shape without tearing.

9.11 Filler

In rigid PVC extrusion the use of relatively large quantities of inorganic powders to reduce costs cannot be applied. Relatively small amounts of filler are used and certain properties, notably impact strength, are improved.

9.12 Level of addition

It is common practice to base this on 100 parts by weight of PVC polymer; frequently termed "parts per hundred resin" or shortened to phr. The advantage of this is that if the level of one additive is increased the figures describing the others remain unchanged.

9.13 Processing terms

"Compounding" describes the conversion of a PVC polymer with additives to a melted stage and then converts this to granules (e.g. as 3 mm cubes) which are cooled and then fed immediately or at a later stage to a final processing machine e.g. an extruder conversion to granules can be made by a "die face cutter" which performs directly on the end of the extruder or by a granular which slits and chops after cooling. The "converter" is a plant which transforms either polymer or granules to the "finished product". Compounding can be done by the converter in which case it is done "in-plant" or prior to delivery to the converter either by the "polymer supplier" or by an intermediate "compound manufacturer".

The "extrudate" describes the shaped melt which issues from an extruder die (e.g. pipe, sheet, rod profile).

"Take-off" equipment describes the ancillary machinery which shapes, cools, conveys, cuts or coils the extrusion. The "haul-off" grips the extrusion and pulls it away from the extruder at a constant set speed.

The "die" at the end of the barrel shapes the melt to the required shape and due to its restriction to melt flow develops a pressure against which the extruder must "pump" the melt. The final section which has an aperture of the required product cross section normally has a parallel "land". The ratio of the length of the aperture to the depth is termed the land length ratio, and can vary from 4:1 to 40:1. The higher this ratio (for a given depth) the higher the pressure developed in the die.

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The staff of RAPRA Process Laboratory for their assistance in carrying out the experimental work.

11. APPENDIX

11.1 High speed mixing.

The mixer used was an 8 litre total capacity model TLHK8 high speed mixer manufactured by Papenmeier having rotor speeds of 3000 and 1500 rpm. The high speed was used for these experiments. Although a heating jacket was fitted this was not used and all mixes were started from ambient temperature. The mixer was fitted with a deflector plate carrying a thermocouple probe, but a second probe having a faster response was used, made from spot welded thermocouple wires enclosed in an epoxy resin impregnated glass fibre sleeve with the junction exposed to the material being mixed. Batch sizes were based on 2 Kg of polymer and cooling was carried out separately in a Hobart planetary mixer.

11.2 Extrusion

This was carried out on a Bone-Cravens 1½" single screw extruder with screw dimensions as follows:-

Diameter 38 mm (1½"). Length 915 mm (36")
Length/Diameter ratio 24:1. Pitch 38 mm (1½")

Screw 1 (3:1) Length of feed zone. 8D. i.e. 305 mm (12")
Length of compression zone 8D i.e. 305 mm (12")
Length of metering zone 8D i.e. 305 mm (12")
Width of flight land 6.3 mm

Screw 2 (2:1) Continuously reducing depth from feed zone to screw tip

	Depth of feed zone.	Depth of metering zone.	Depth ratio.
Screw 1	10.6 mm (.405")	5 mm (.196")	2:1
Screw 2	12.6 mm (.495")	4.2 mm (.165")	3:1

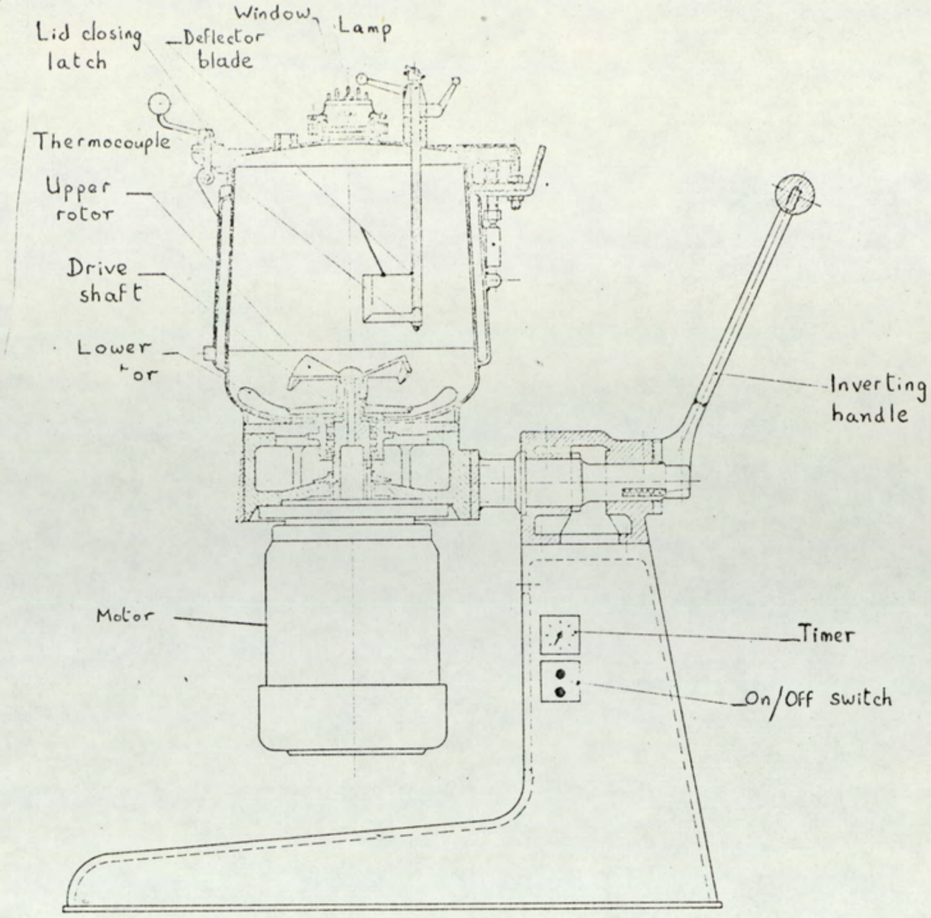


Fig. 68 Papenmeier TLHK 8 laboratory high speed mixer

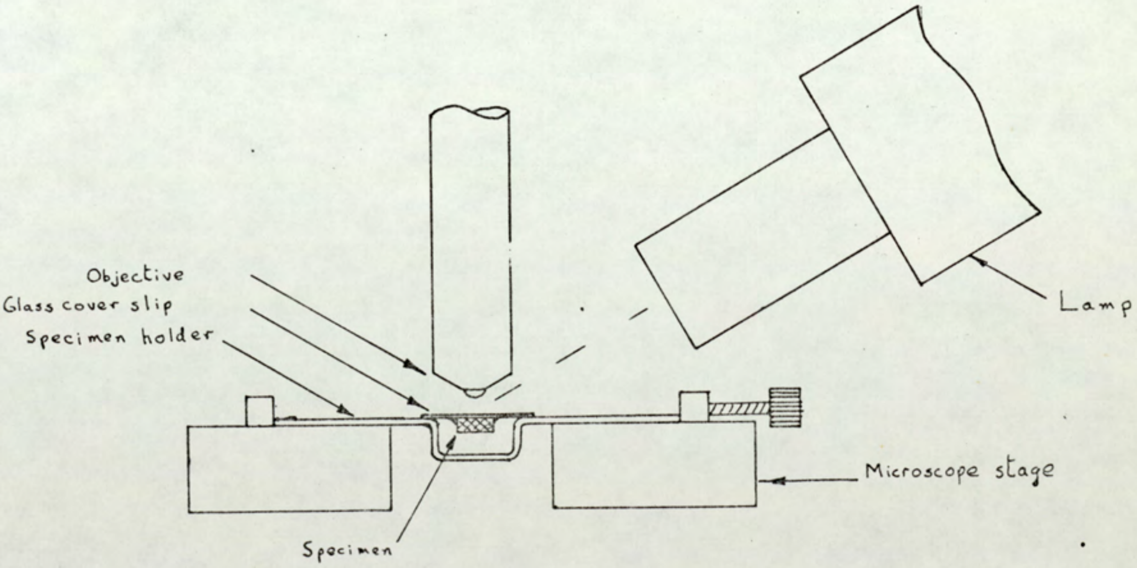


Fig. 69 Method for mounting specimens on microscope stage

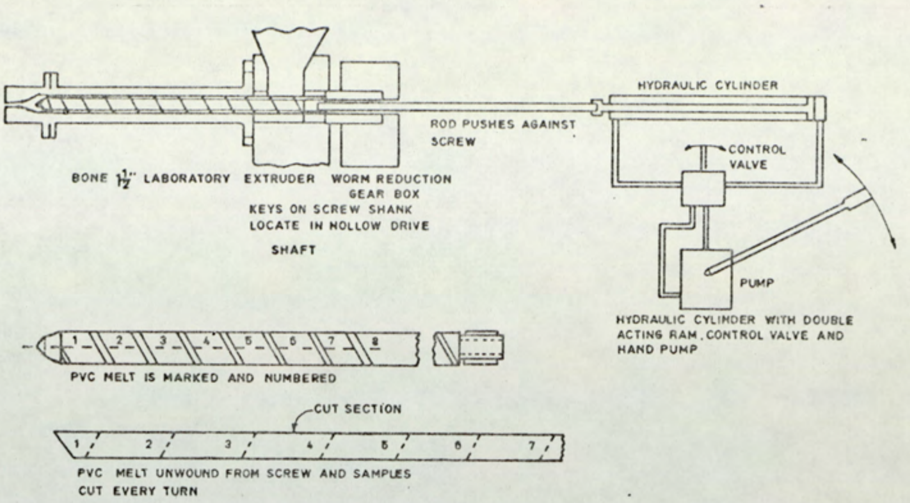
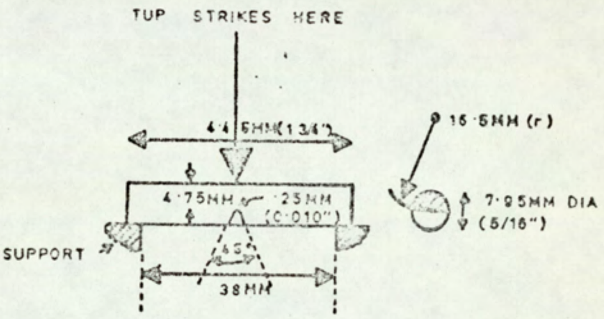


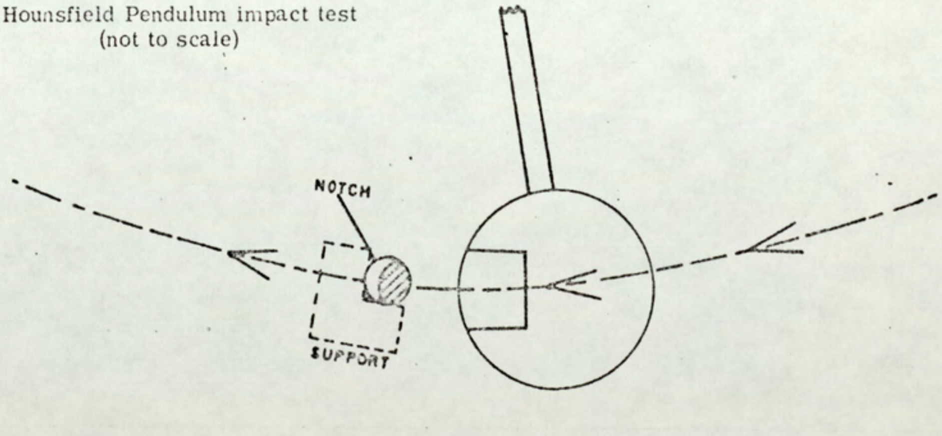
Fig. 70 Hydraulic jack for removal of extruder screw

Fig. 71 Impact test

Notched Impact Bar details



Hounsfield Pendulum impact test
(not to scale)



Speed range 0-110 continuously variable.

Motor power 7.5 HP

Heating by electrical resistance heaters, three zones on barrel, one or two on die. Cooling by "Willert" vapour cooling system.

Screw removal was by using a double acting 2" hydraulic cylinder pushing a bar against the screw shank via a hollow drive shaft. Hydraulic pressure was applied from a hand pump via a four way valve. Maximum available load was 2 tons. As the stroke was limited to 600 mm (24"), in a number of cases it was necessary to return the double acting ram and insert a second bar before continuing with the screw removal. In most cases the screw could be removed completely from the extruder within two minutes of stopping screw rotation. For such experiments simple rod dies were used as quick easy removal was necessary with easy cleaning. Cleaning of the die was necessary immediately after removal to avoid thermal degradation.

Die (1) 8 mm ($5/16"$) diameter rod to fit Tensometer impact tester.

Die (2). Electrical conduit profile die. This produced a square channel 25 mm wide by 13 mm deep with a wall thickness of 2 mm. It had various lugs for wall fixing and attaching a clip-on cover.

11.3 Impact Testing

Notched impact bar tests were carried out using a Charpy type of pendulum machine made by Tensometer Ltd. The extruded rods 8 mm (5/16") dia were cut into 44 mm ($1\frac{3}{4}$") lengths and notched on a Tensometer notching machine. Twenty specimens were then impact tested. The channel sections were cut into 152 mm (6") lengths and tested on a falling weight machine using the probit method.

11.4 Microscopy

PVC specimens were prepared by filing flat the sawn edges and wet sanding with "wet or dry" (W.O.D) paper starting at 220 grade and working down through finer grades, 320, 400 and 500, finally polishing with "Belco" rubbing compound. The polished end was then cut off to give a specimen 3 mm thick. This specimen was then put in a glass specimen tube which was filled with hydrogen sulphide gas, stoppered and left overnight. This converted the lead stabiliser to lead sulphide, which being an intense black enabled easy identification of stabiliser particles and therefore outlines of unfused PVC particles.

The specimens were then mounted on the microscope stage by adhering the polished face to the underside of a glass coverslip with Lenzol (cedarwood oil). This was a suitable mounting liquid as it has a refractive index similar to that of PVC, while its surface tension was sufficient to hold the specimen in contact with the cover slip. The coverslip was then placed on a fabricated aluminium holder having the same overall size as a microscope slide. The design was such that sufficient movement on the microscope stage was possible for examination of the whole of the specimen surface.

Illumination was by reflected light using a microscope lamp shining obliquely on the specimen. Illumination was insufficient for use at high magnification, but in general lower magnifications were of greater interest as larger areas of channel cross section could be viewed, showing distribution of unfused PVC. 32 mm and 16 mm objectives were mainly used.

For measurements of fusion rate, the degree of fusion at a particular point along the screw was taken as the proportion of the cross sectional area occupied by the fused material, (expressed as a percentage). The most convenient procedure was to use one side of a steoreo binoccular microscope fitted with a "Camera Lucida", which is a prism device placed on the eyepiece enabling a drawn outline to be seen as a second image superimposed on the specimen. Measurement of the area could then be made either by using a planimeter or by weighing outlines cut from thin card. The latter method was found to be quicker and more accurate than the planimeter, while use of photomicrographs proved difficult and slow.

11.5 Hopper flow measurements

These were carried out to the method of ASTM D1895-65T. The apparatus consisted of a stainless steel hopper 114 mm high by 93 mm diameter at the top with a discharge aperture at the bottom 1 mm dia. The procedure was to fill the hopper with 100 g of powder with the aperture closed by a finger and to measure the time taken for the hopper to empty.

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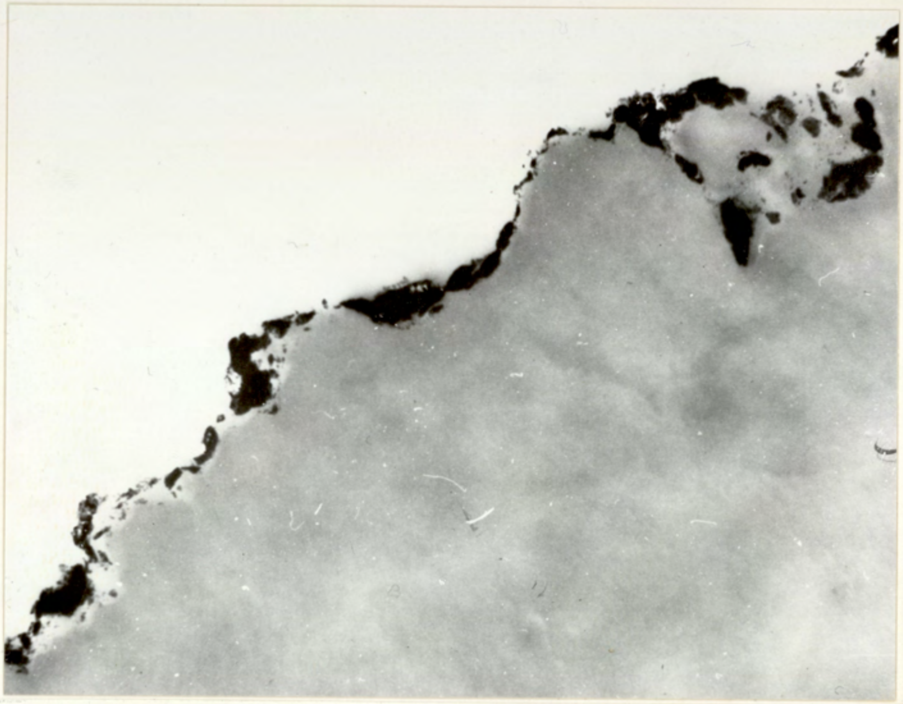
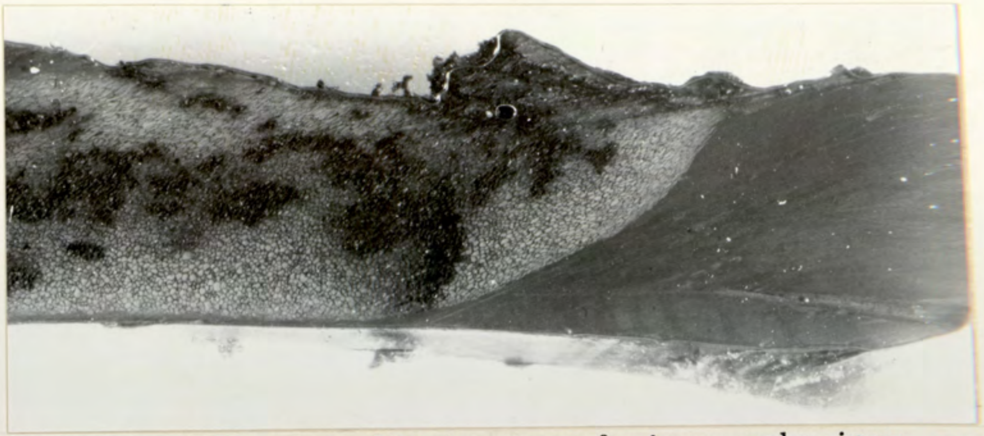


1. Electronmicrograph showing coating of lead stabiliser on PVC particle.

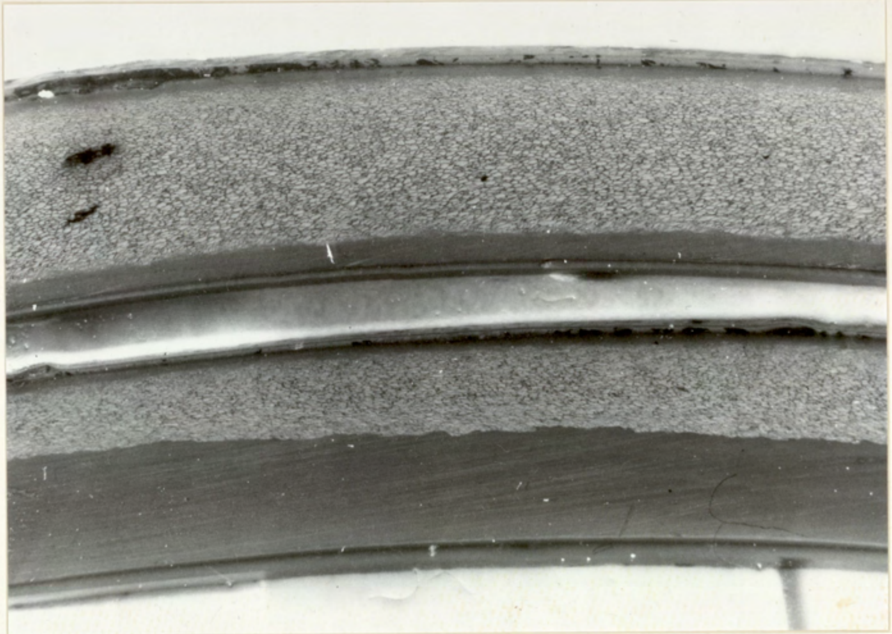


2. Screw cross channel section showing rear fusion mechanism — fusion just started.

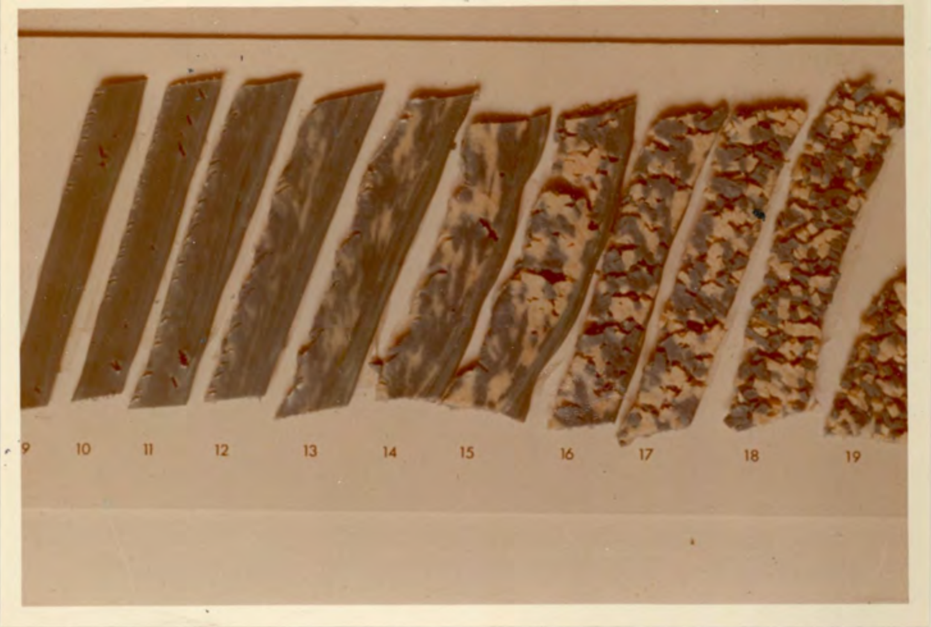
3. Screw cross channel section showing rear fusion mechanism - intermediate stage.

4. Screw cross channel section showing rear fusion mechanism — fusion nearly completed.

5. Screw channel cross section showing entrapped air and patches of unfused PVC.



6 Screw down channel section showing rear fusion mechanism - intermediate stage.

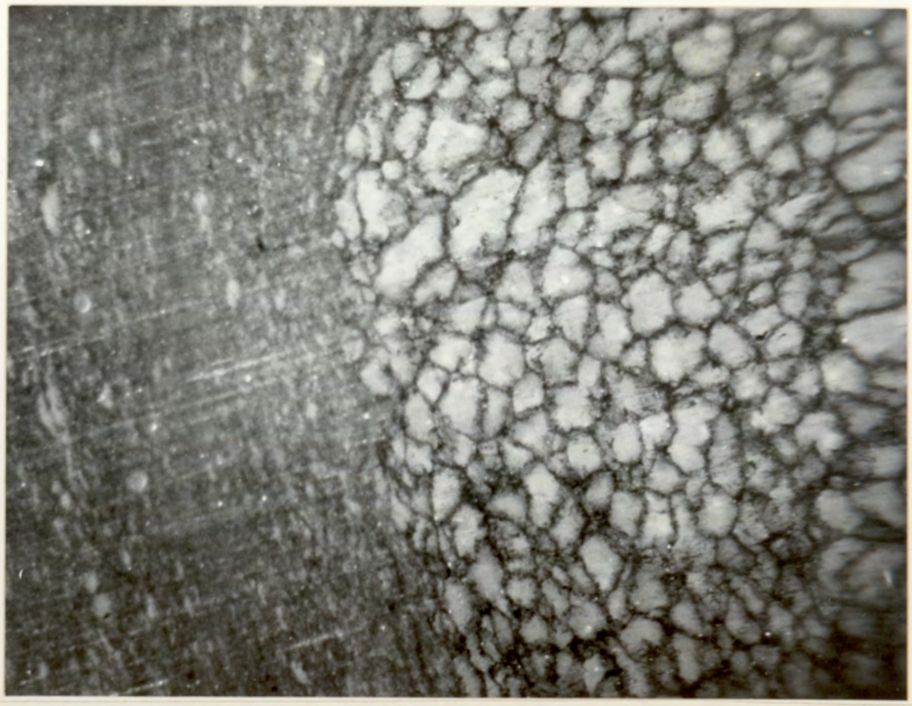


7. Material removed from screw channel - rear fusion mechanism with PVC granules.

8. Material removed from screw channel - rear fusion mechanism with coloured PVC dryblend.

9. Material removed from screw channel - rear fusion mechanism with PVC dryblend containing coloured marker granules.

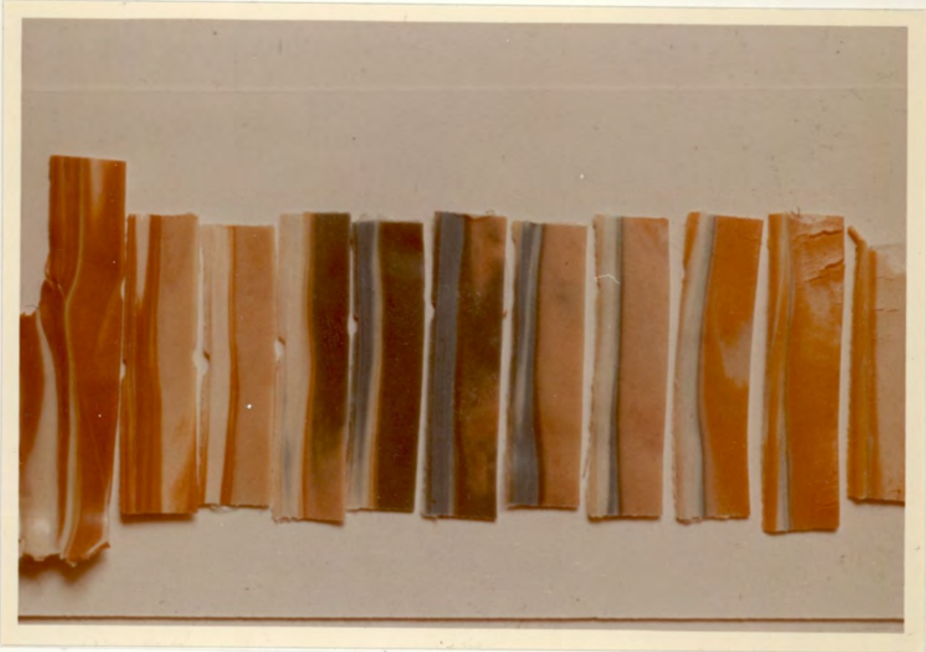
10. Screw cross channel section showing forward fusion mechanism with Solvic VP 17/10 (above) and PVC dryblend containing 1 phr Wax E. (below)



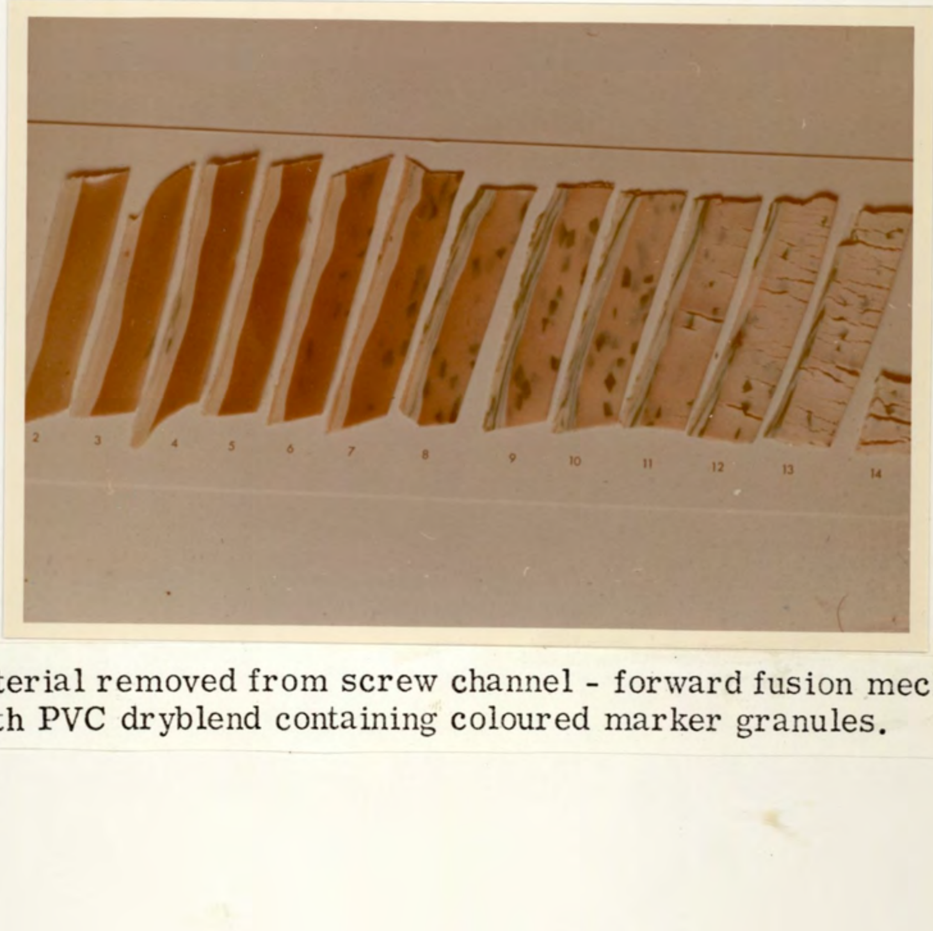
11. Screw cross channel section showing forward fusion mechanism with 1 phr Wax E.

12. Screw cross channel section showing forward fusion mechanism with Solvic VP 17/10 dryblend.

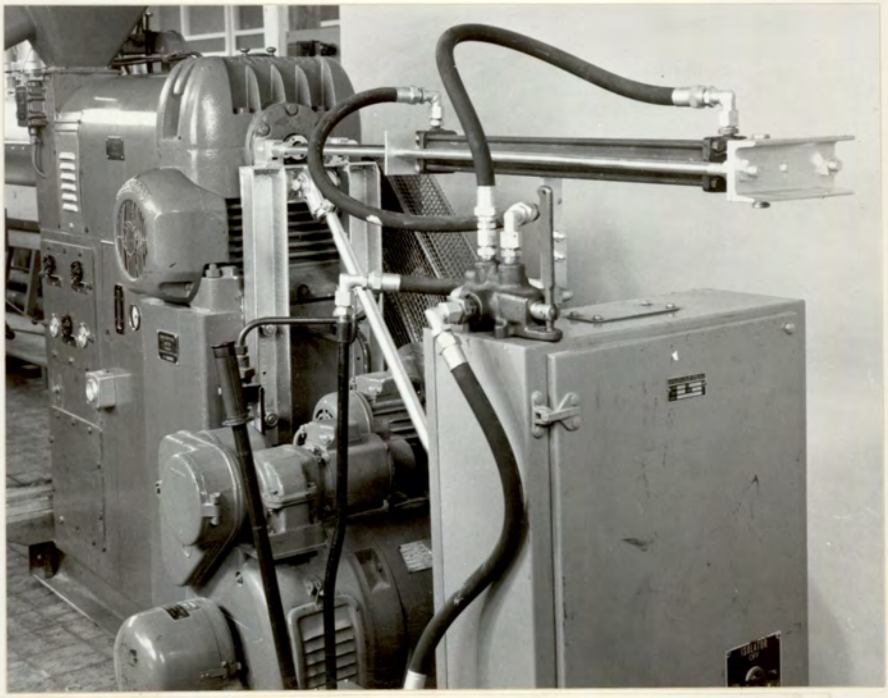
13. Screw down channel section showing forward fusion mechanism.



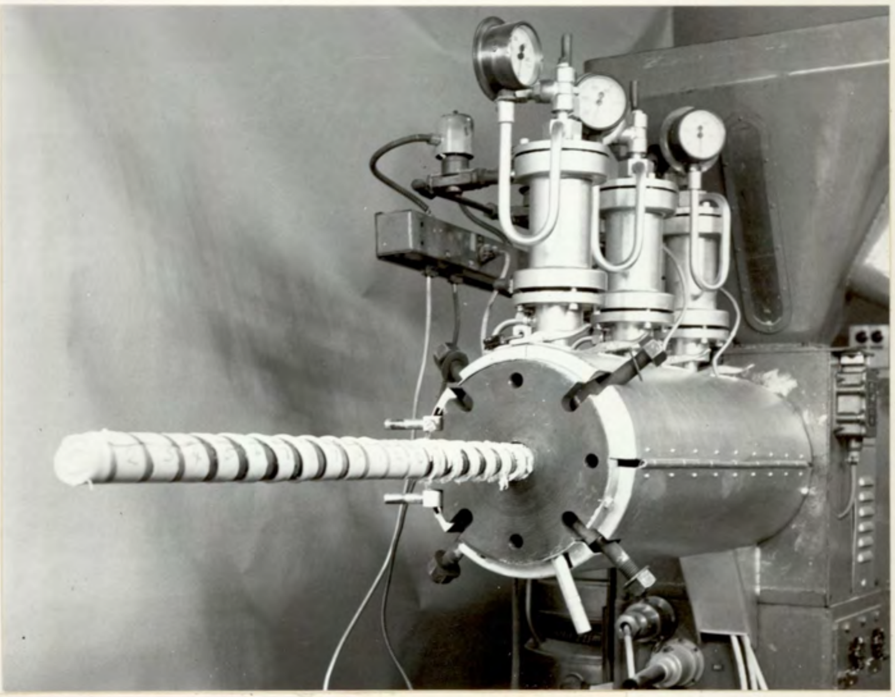
14. Material removed from screw channel - forward fusion mechanism with coloured dryblend.



15. Material removed from screw channel - forward fusion mechanism with PVC dryblend containing coloured marker granules.



16. Extruder, showing hydraulic ram for screw removal.



17. Extruder, showing screw extended