# Drawing of polymers through a conical die

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A new process is described in which ultra-oriented polymers are produced by drawing a billet of initially isotropic polymer through a converging die. The process, called die-drawing, has been used to make oriented polypropylene rods with room temperature Young's moduli up to 20.6 GPa. The considerable advantages of this process compared with conventional tensile drawing and hydrostatic or ram extrusion are discussed.

## INTRODUCTION

Techniques have recently been reported for the production of ultrahigh modulus oriented polymers in the form of fibres or rods. Conventional tensile drawing, either of compression-moulded specimens on a tensile testing machine[1–3] or of melt-spun filaments by a continuous process[4,5] has been employed to obtain products with typical diameters of ~ 1 mm for circular sections. Hydrostatic extrusion[6–9] has allowed production of rods up to ~ 25 mm diameter. Both of these solid phase deformation techniques produce materials with an axial Young's modulus in the range of glass or aluminium for linear polyethylene (LPE).

Conventional tensile drawing processes have certain limitations. Drawing on a tensile testing machine (*Figure 1a*) is a batch process with a considerable waste of material in the grips, and severely limited production lengths. Because of practical limitations, this method is not readily applicable for sections greater than, say, 10 mm original diameter. Furthermore, if drawing is performed at elevated temperatures, deformational heating of thick specimens can lead to failure. Drawing at lower temperatures incurs higher drawing stresses, and, consequently, gripping problems. Continuous drawing of filaments (*Figure 1b*) is also limited in practice by the initial diameter of the isotropic filament which can be processed: diameters up to ~ 2 mm are generally employed.

Hydrostatic extrusion (*Figure 1c*) and ram extrusion have been explored as a means of producing larger bulk specimens than can be obtained by conventional tensile drawing processes, and rods with diameters in the range ~ 2 to ~ 25 mm have been successfully produced[6–11]. However, hydrostatic extrusion suffers from certain drawbacks which can in general be only partly overcome. First, hydrostatic extrusion is a batch process, requiring a discrete billet to be loaded into a temperature-controlled pressure vessel. Secondly, the strain rate field imposed by the process is usually detrimental to fast and in the case of higher deformation ratio products to practical–production rates under isothermal conditions[6–9,12]. The high pressures involved in hydrostatic extrusion also inhibit the process, drastically in some cases, e.g. polyoxymethylene (POM)[9]. Consequently, certain polymers, notably polypropylene (PP), intermediate molecular weight LPE and POM can only be extruded to deformation ratios, $R$

$$\left(\text{where } R = \frac{\text{initial cross sectional area}}{\text{final cross sectional area}}\right)$$

considerably lower than those which can be attained by tensile drawing e.g. $R \sim$ 0 for POM, $R \sim 7$ for PP, compared with draw ratios of ~ 20 for tensile specimens. Attempts have been made to circumvent these difficulties in hydrostatic extrusion. Extruding under adiabatic conditions has met with some success in LPE[11] but does not appear to be feasible for, say, PP or POM. Application of a tensile haul-off force to the extrudate ('haul-off assisted extrusion', *Figure 1c*) is known to improve extrusion pro-

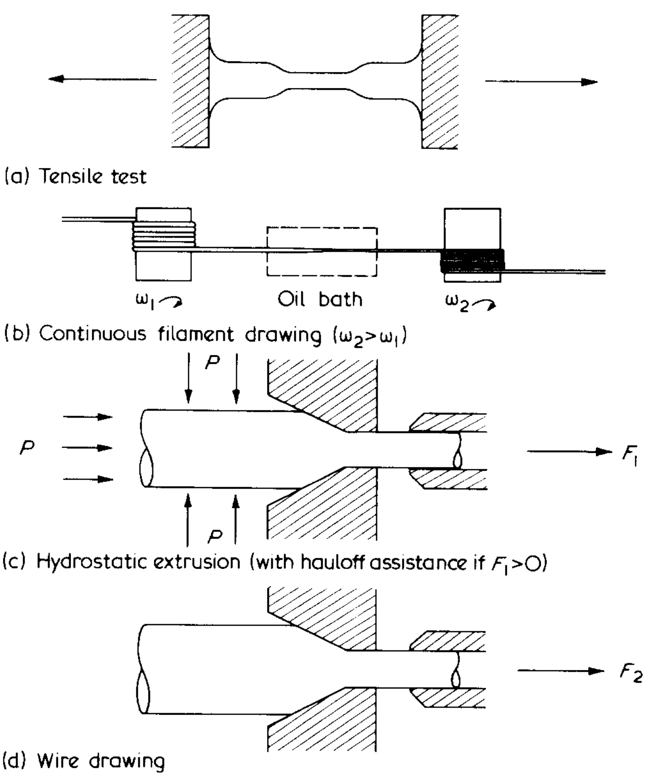


*Figure 1* Some solid phase deformation processes

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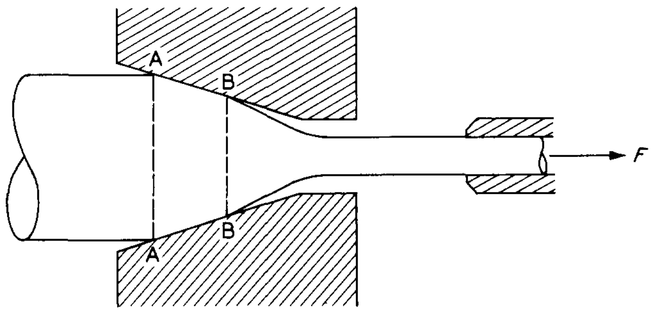


Figure 2 Die drawing process. Isotropic material encounters die at AA; deforming material ceases to be in contact with die at BB

duction rates, but this effect has only been small at the higher extrusion ratios since haul-off stresses have generally been low compared with the extrusion pressure. The application of increasingly larger proportions of haul-off stress to extrusion pressure, mainly to counteract any tendency of the extrudate to swell on leaving the die, has also been investigated[13].

The aim of the present work has been to obtain high stiffness oriented rods by a process which avoids the limitations found in hydrostatic extrusion and conventional tensile drawing as outlined above. This has been achieved by the tensile drawing of the polymer through a die, and will be referred to as 'die drawing'.

## DIE DRAWING PROCESS

The principle of the die drawing process is illustrated in *Figure 2*. An axial tensile force, $F$, is applied to a heated solid polymer billet. As $F$ is increased from zero there are several possible outcomes, depending on the billet material, and the process conditions. The major process variables are the billet–die geometry, the billet–die temperature, the magnitude of the tensile force and the rate of drawing. In extreme cases, on application of the tensile force there may occur: (i) brittle failure at the die exit; or (ii) the situation where the die behaves purely as a clamp, so that a simple tensile test ensues. For successful die drawing, application of the tensile force leads to steady drawing of the polymer through the die at some diameter smaller than the die exit bore diameter, the polymer deforming in such a manner that it is no longer in contact with the die wall throughout the die (i.e. the polymer 'necks' down). Under suitably chosen drawing conditions it is well known that the neck formation in certain polymers can be stabilized by high strain hardening behaviour[4,14]. It is therefore possible to envisage 'steady state' drawing conditions for the die drawing process, provided that the polymer strain hardens suitably and appropriate values of process variables are employed.

It is clear that if a large deformation ratio is attempted the die will behave purely as a clamp. It was therefore originally considered that an oriented initial 'nose' of polymer must pass through the die. Because this nose would possess a higher yield stress than the isotropic material it could therefore sustain a tensile force of sufficient magnitude to deform the isotropic material in the die, thus allowing steady drawing of the billet through the die.

An intermediate case, not previously described elsewhere, is also plausible: it is conceivable that a process identical to that of wire drawing *Figure 1d* may be operated. In this process, the polymer will be drawn through the die, remaining in contact with the wall throughout the die, to form a die exit diameter product.

## EXPERIMENTAL

### *Materials*

The material investigated was a polypropylene copolymer, 'Propathene' GSE 108 (Melt Flow Index, *MFI*, 0.8) manufactured by ICI Ltd. Polypropylene is a particularly good material to investigate by die drawing since: (a) it is commercially available in rod form; (b) it is capable of being drawn to very high draw ratios, with an accompanying increase in Young's modulus, hence making a desirable product[3,15], and (c) the unassisted hydrostatic extrusion behaviour of polypropylene has been found to be limited to deformation ratios of about 7 (although haul-off assisted extrusion allows higher deformation ratios to be attained, with considerably increasing difficulty[10,13,16]). This value is drastically lower than the previously reported attainable draw ratios: die drawing was therefore envisaged as a practical means of production of high stiffness large diameter polypropylene rods.

### *Apparatus*

Die drawing experiments have been conducted with two different experimental arrangements. In the first instance a small scale rig (*Figure 3*) was constructed for use with an Instron Tensile Testing machine. The rig was designed to allow small initial billets to be die drawn. This is a useful feature if the polymer is not commercially available in rod

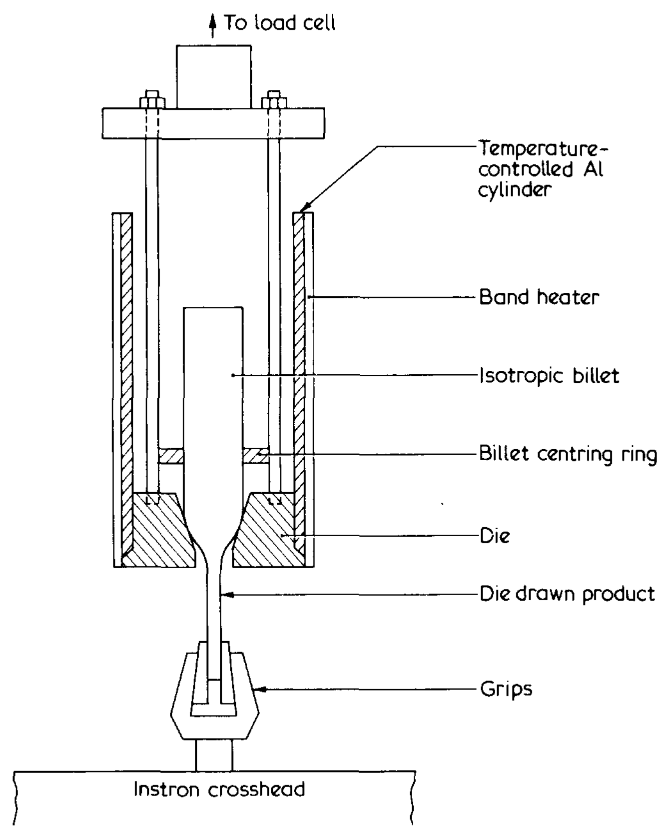


*Figure 3* Small scale die drawing rig (mounted on Instron Tensile testing machine

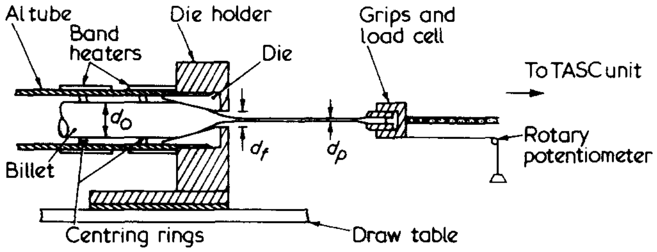


*Figure 4* Large scale die drawing rig

form and, consequently, has to be compression-moulded or injection-moulded. The rig also allows a comparison of results with those obtained from a larger rig, indicating any scaling-up effects.

The conical die had a 15° semi-angle and 7 mm diameter exit bore, and was suitable for use in a hydrostatic extrusion vessel described in ref. 8, for production of an oriented initial nose if so desired. The die assembly was temperature controlled to an accuracy of ± 1°C. A set temperature of 110°C was employed throughout the experiments because previous studies showed this to be in the optimum drawing temperature range for polypropylene[3].

The second rig (*Figure 4*) was of larger scale, employing a 15.5 mm exit bore diameter conical die which again had a 15° semi-angle. This rig is designed for horizontal operation, over a length of ~ 7 m and is similar to a conventional draw-bench. The die assembly was temperature-controlled to an accuracy of ± 2°C. A set temperature of 110°C was employed throughout the experiments. The large diameter billets (up to ~ 40 mm) were preheated before loading into the die assembly. The draw force was provided by a Pye TASC (Torque and Speed Control) unit, and was measured by a load cell in the grip assembly. The grip assembly motion was monitored using a calibrated rotary potentiometer system. The die was again suitable for use in the hydrostatic extrusion vessel. Replacing the die assembly by this extrusion vessel provides in effect a third die drawing rig which can be directly employed if an extruded initial tag is used, although the extrusion vessel clearly limits the initial billet length.

For a given die, the choice of a particular billet diameter $d_0$ will fix the nominal deformation ratio, $R_N$:

$$\text{where } R_N = \frac{\text{Original billet cross-sectional area}}{\text{Die exit cross-sectional area}} = \left(\frac{d_o}{d_f}\right)^2$$

However, since the product is expected to be of smaller diameter than the die exit bore, the *actual* deformation ratio, $R_A$, is given by:

$$R_A = \frac{\text{Original billet cross-sectional area}}{\text{Final product cross-sectional area}} = \left(\frac{d_o}{d_p}\right)^2$$

$R_A$ corresponds to the conventional draw ratio, λ, assuming deformation occurs at constant volume.

## *Experimental procedures*

*Formation of oriented nose.* Two methods have been used in the present work to form a nose which has a sufficiently high yield stress to enable the desired nominal deformation ratio billet to be drawn through the die. The first method involved hydrostatic extrusion of a nose of $R_N \sim 5$ at 110°C in order to die draw a billet of nominal deformation ratio 5 or 7. The second method involved a drawing-only technique, not previously considered feasible. This technique employs a stepped billet, i.e. a billet having steps in diameter along its length (*Figure 5*). The initial nose is isotropic and of a slightly smaller diameter than the die exit bore diameter, $d_f$. The first stage of the billet has diameter $d_1$, chosen to give a sufficiently low value of nominal deformation ratio, $R_{N_1}$ to allow drawing of this section through the die by pulling on the isotropic nose. If drawing is performed at low speeds, this may indeed be achieved. Some necking may occur, but the load bearing ability of the polymer will be maintained provided that it strain-hardens sufficiently. Once the neck has stabilized and is moving axially away from the die into a zone of lower temperature and therefore cooling, the product diameter at the die exit approaches $d_f$ again. It is clearly possible to enhance the load-bearing ability of the nose by cooling e.g. by means of water at ambient temperature, and this has been employed in some cases where the neck might not have stabilized unless cooled.

Having drawn the first stage of the billet through the die the now oriented product (of deformation ratio $\simeq R_{N_1}$) can be gripped and the process repeated for the second stage of the billet, again at low draw speed. The product of deformation ratio $\simeq R_{N_2}$ so obtained may then be gripped, in order to draw the maximum nominal deformation ratio stage through the die. Slow drawing of this final stage produces a nose suitable for the desired die drawing. This process therefore involves only drawing of the stepped isotropic billet, moving the clamps along the product at each stage. The number of stages depends on the desired maximum $R_N$. For example for $R_N = 5$ it has been found practical to employ a first stage with $R_{N_1} = 2$ to 3, followed by the $R_N = 5$ billet. For $R_N = 7$, two stages $R_{N_1} = 2$ to 3, $R_{N_2} = 5$ have been used. The initial stages need only be of a length sufficient to obtain material which can be gripped.

*Drawing to high deformation ratios.* A similar procedure was followed for both small and large scale die drawing. When an oriented nose had been produced by either of the two methods described above, the nose was gripped, a constant drawing speed, *v*, was set, and the drawing load was monitored. Since the final draw ratio was found to depend on the drawing speed (see below) each billet was only used to obtain one sample at one value of drawing speed, *v*. This clearly restricts the collection of experimental data compared with, say, hydrostatic extrusion, where many extrusion

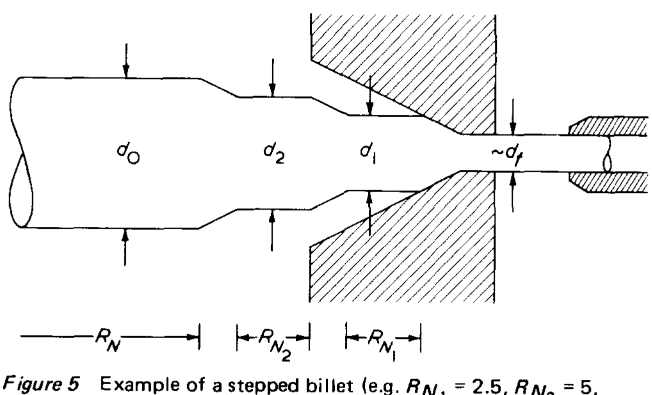


*Figure 5* Example of a stepped billet (e.g. $R_{N_1} = 2.5$, $R_{N_2} = 5$, $R_N = 7$)

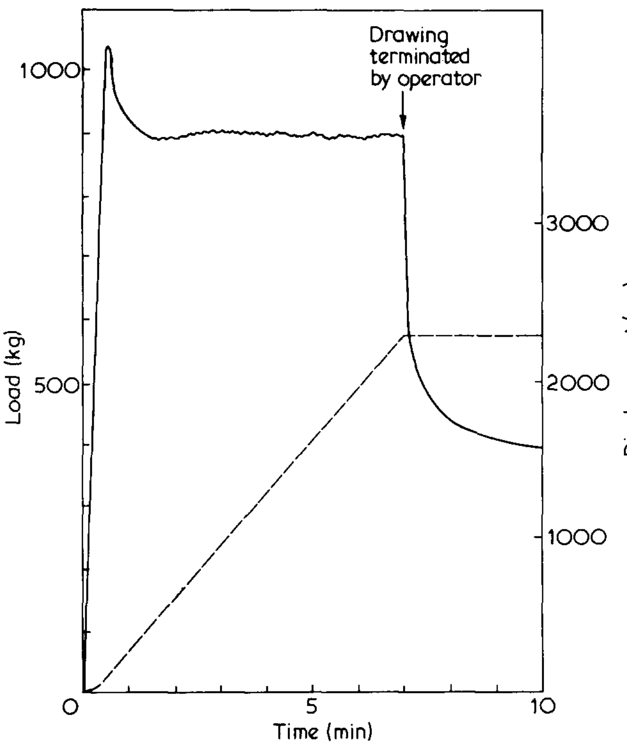


*Figure 6* Typical die drawing load–time (—) and displacement–time (— — — —) curves for PP copolymer nominally at 110°C. (Actual sample $R_N$ = 7)

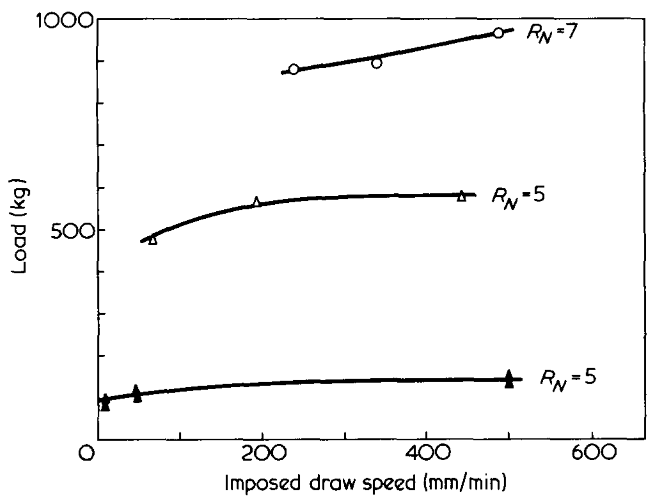


*Figure 7* Relationship between steady state draw load and imposed draw speed for PP copolymer nominally at 110°C. (O, $R_N$ = 7, 15.5 mm die; Δ, $R_N$ = 5, 15.5 mm die; ▲, $R_N$ = 5, 7 mm die)

pressure–extrusion speed data can be obtained from a single billet.

The small scale rig was strictly limited in the length of sample which could be produced at the final draw ratio for a given drawing speed, whereas drawing on the large scale rig had to be terminated when practically manageable lengths of the final draw ratio product had been obtained (i.e. ~ 3 m).

## RESULTS

Both the extruded nose and the drawing only techniques have been employed successfully, and products up to draw ratio 20 with Young's modulus up to 20.6 GPa have been obtained. In the drawing-only process it was found that drawing of the initial deformation ratio sections had to be limited to a rate of about 10 mm/min (without forced cooling of the nose) to avoid failure of the product. Even with forced cooling (by water) a rate of 10 mm/min was chosen for these initial stages.

The following results refer to die drawing of the maximum nominal deformation ratio section of the billet. *Figure 6* shows a typical load–time curve and a typical displacement–time curve for the die drawing process. The load–time curve exhibits a maximum in the initial stage of drawing, followed by a steady load plateau, which persists for the length of the driving experiment. A steady velocity is maintained in the experiment which is, of course, equal to the imposed constant velocity.

The variation of steady load with imposed draw speed is shown in *Figure 7*, the load being the dependent variable, for billets having a maximum $R_N$ of 5 or 7. In both cases the load rises slowly with draw speed.

Because drawing occurs in the presence of a neck at all times the drawing stress will vary with location along the product. We can, however, define two useful parameters, the maximum draw stress, which occurs at the maximum draw ratio and the minimum draw stress, corresponding to initial yield. The rate of deformation will, of course, be different for these two quantities, the higher deformation ratio material deforming at the slower rate. Bearing these points in mind, the variation of maximum and minimum stress with imposed draw speed are shown in *Figure 8*. The maximum draw stress, $\sigma_{max}$, rises markedly with increasing

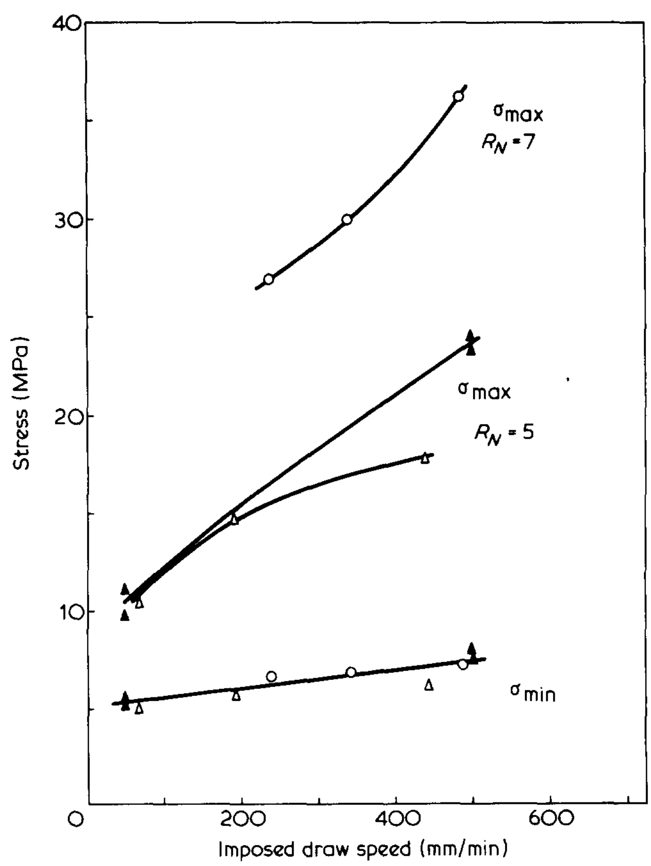


*Figure 8* Maximum and minimum draw stress along a specimen *versus* imposed draw speed, for PP copolymer nominally at 110°C. (O, $R_N$ = 7, 15.5 mm die; Δ, $R_N$ = 5, 15.5 mm die; ▲, $R_N$ = 5, 7 mm die)

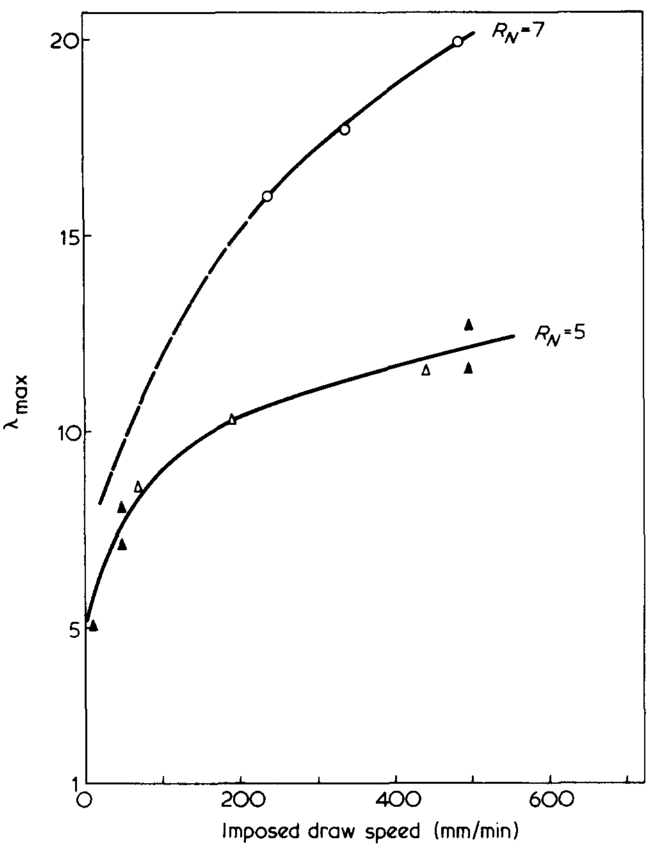


*Figure 9* Dependence of maximum steady draw ratio, $\lambda_{max}$, on imposed draw speed for PP copolymer die drawn at a nominal temperature of 110°C. (O, $R_N$ = 7, 15.5 mm die; Δ, $R_N$ = 5, 15.5 mm die; ▲, $R_N$ = 5, 7 mm die)

draw speed, $v$, especially in the case of $R_N$ = 7 billets, where the rate of increase of $\sigma_{max}$ increases with $v$. The minimum stress values, $\sigma_{min}$, lie close together, roughly on the same curve. This is to be expected since the $\sigma_{min} - v$ curve is, in effect a plot of the isotropic material yield stress against strain rate and should not depend upon the nominal deformation ratio. In general small scale drawing appears to incur slightly higher values of $\sigma_{max}$ than large scale drawing. This may be related to deformational heating effects, which are discussed later.

The relationship between maximum *steady* draw ratio, $\lambda_{max}$, and the imposed draw speed, $v$, is shown in *Figure 9*. This is a particularly useful representation of die drawing results for production purposes, as well as having intrinsic scientific interest. The maximum steady draw ratio clearly increases with increasing draw speed, the rate of increase being considerably greater for the $R_N$ = 7 billets.

Finally, the products were evaluated by optical inspection and by measurement of the axial Young's modulus. All products except $\lambda$ = 19.9 had a very smooth surface and were quite transparent. The $\lambda$ = 19.9 product was opaque white, possibly due to internal voids, and exhibited surface fibrillation on bending. The straightness of the product was found to be strongly dependent on the billet being drawn along its axis, rather than at an angle to its axis: initial billets were drawn without the billet centring rings (*Figures 3* and *4*) resulting in a curved product. The centring rings enabled straight rods to be produced. The axial Young's modulus was measured at ambient temperature in a three point bend test, using a support separation of 1 m, to obviate end effects. The application of this test to anistropic polymers has been described elsewhere[17]. The variation of Young's modulus at 0.1% maximum (surface) strain with $\lambda_{max}$ is shown in *Figure 10*. Large increases in Young's modulus over the isotropic material (where $E \simeq 1.1$ GPa[18]) were obtained. There appears approximately to be a linear relationship between modulus and draw ratio up to $\lambda_{max} \sim 13$, above which the slope of the curve decreases. For comparison, *Figure 10* also shows the results of Cansfield *et al.*[3] for PP homopolymer fibres drawn at 110°C, and those of Williams[10] for PP homopolymer rods hydrostatically extruded at 100°C. There is generally excellent agreement, within the experimental error, between these results. For convenience, a summary of the die drawing results is presented in *Table 1*.

## DISCUSSION

### *Die drawing process*

The preceding results clearly show that die drawing is a very successful method for the manufacture of high stiffness PP rods. Recalling some of the disadvantages of other solid phase deformation processes discussed above, it is now valuable to assess the die drawing process.

*Production rates.* First, the die drawing process unlike hydrostatic extrusion does not suffer from the disadvantage of decreasing production rates at increasing deformation

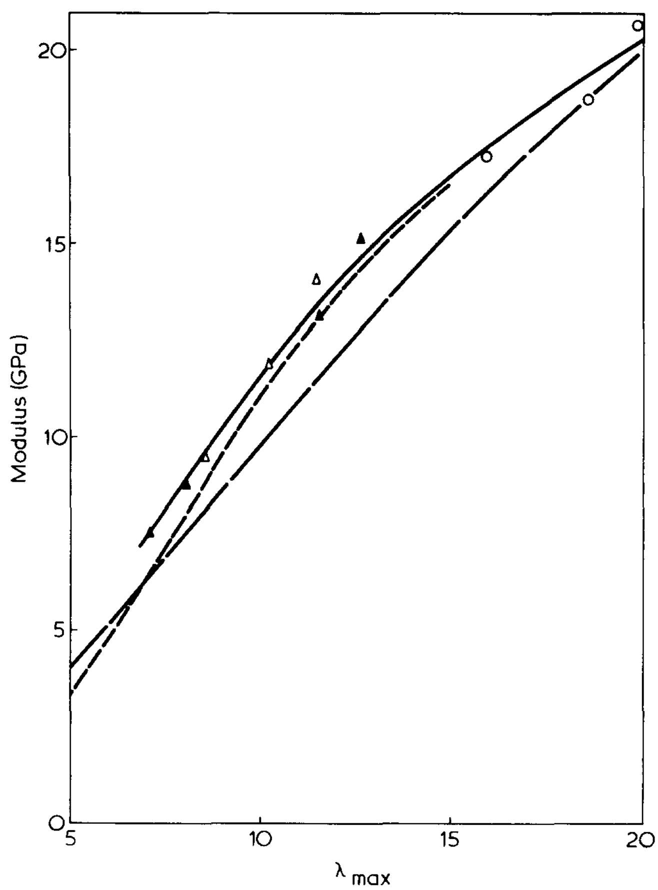


*Figure 10* Axial Young's modulus, determined at 0.1% maximum strain at 21°C, *versus* maximum steady draw ratio for PP copolymer die drawn at a nominal temperature of 110°C. (O, $R_N$ = 7, 15.5 mm die; Δ, $R_N$ = 5, 15.5 mm die; ▲, $R_N$ = 5, 7 mm die.) The results of Cansfield *et al.*[3] for PP homopolymer fibres drawn at 110°C, (—— ——)and Williams,[10] for PP homopolymers hydrostatically-extruded at 110°C (– – – –) are included for comparison

*Table 1*

| $R_N$ | Die bore (mm) | Steady load *L (kg)* | Product minimum steady diameter (mm) | $\lambda_{max}$ | Draw speed *v* (mm/min) | Young's modulus for $\lambda_{max}$ (GPa) |
|---|---|---|---|---|---|---|
| 2 | 7 | 36 | 7 | 2 | 10 | – |
| 2 | 7 | 32.5 | 7 | 2 | 10 | – |
| 5 | 7 | 93 | 7 | 5 | 10 | – |
| 5 | 7 | 92.5 | 7 | 5 | 10 | – |
| 5 | 7 | 113 | 6.00 | 7.1 | 50 | 7.6 |
| 5 | 7 | 110 | 5.55 | 8.1 | 50 | 8.8 |
| 4.7 | 7 | 148 | 4.45 | 11.6 | 500 | 13.2 |
| 5 | 7 | 152 | 4.45 | 12.7 | 500 | 15.1 |
| 5 | 15.5 | 480 | 12.00 | 8.6 | 67.6 | 9.5 |
| 5 | 15.5 | 570 | 11.00 | 10.3 | 144.2 | 11.9 |
| 5 | 15.5 | 576 | 10.09 | 11.5 | 442.9 | 14.1 |
| 7 | 15.5 | 885 | 10.12 | 15 | 238.5 | 17.3 |
| 7 | 15.5 | 900 | 9.65 | 17.7 | 339.9 | 18.7 |
| 7 | 15.5 | 970 | 9.15 | 19.9 | 486.5 | 20.6 |

ratios. In fact the present results indicate quite the opposite—the maximum deformation ratio $\lambda_{max}$ increases with draw speed, i.e. the higher deformation ratio products are obtained at faster rates. This is related to two main features of the mechanics of die drawing: (1) the low hydrostatic stress component compared with hydrostatic extrusion and (2) the strain rate field in the deforming polymer being imposed by the polymer and drawing conditions rather than by the die geometry and extrusion conditions as occurs in hydrostatic or ram extrusion.

The absence of applied hydrostatic pressure in die drawing leads to a relatively low hydrostatic stress component. Hydrostatic pressure has been observed to have a serious limiting effect on the deformation behaviour of crystalline polymers such as LPE, POM and PP[8,9,12,16]. The polymer in contact with the die wall in die drawing will experience a stress normal to the die wall, but this material is in general only of low deformation ratio, i.e. in a region of deformation ratio where pressure effects are relatively small.

The second factor known to limit the extrusion rate is the unfavourable strain rate field encountered in a converging die[12]. This strain rate field is such that the highest strain rates are encountered at the highest levels of plastic deformation, a situation which incurs very high flow stresses as the polymer reaches the die exit, and hence detrimental high extrusion pressures. By contrast, in die drawing the polymer 'necks' i.e. forms a geometrical profile which reflects an optimal strain rate field for the material[14]. This strain rate field, in common with other axial drawing processes, is such that the highest strain rates are encountered at low levels of plastic deformation, consistent with a constant load at all sections of the product. Only the polymer in contact with the die wall surface suffers a strain rate field imposed by the die geometry. Again, since this material is of low deformation ratios where strain rate effects on the flow stress are comparatively small,[4] the overall effect is not detrimental to the process.

Although die drawing might be considered unlikely, in general, to be employed for production of fibres, the production rates for a continuous die drawing process and conventional fibre drawing should be similar.

*Continuous process possibilities.* The work described in this paper has been concerned with a batch process. However, it is quite feasible that the die drawing process could be operated on a continuous basis, an oriented nose once having been formed. The potential for a continuous process is due to the polymer billet deforming through the die from an essentially open vessel, i.e. the drawing equipment does not impose a maximum length on the billet as, say, an extrusion pressure vessel does. For suitable polymers the billet could be melt extruded in line with the drawing rig (requiring accurate control of the screw extruder[19]). Material wastage, which is economically important, would thus be greatly reduced.

*Stability and control of the process*

Stability and control of a process imply the ability to obtain a desired product. In hydrostatic extrusion the product dimensions and the deformation ratio are fixed mainly by the billet–die geometry, and products can be obtained to close tolerances. Since die drawing involves the presence of an instability (the neck) it might at first be considered unsuitable for the production of a controlled product. However, this is not the case. The results of the present work show that a 'steady state' drawing situation is reached, in which drawing at some imposed steady speed takes place under constant load conditions, and a steady value of $\lambda_{max}$ is obtained. This is rather like the draw load reaching a steady value in a typical test while the neck propagates: in die drawing, the neck is always propagating. The steady state deformation can only be attained in polymers which exhibit a sufficiently high strain hardening behaviour to stabilize the neck. In such polymers strain hardening should allow stabilization over a wide range of drawing temperatures and speeds. A further factor possibly aiding stability in the die drawing process described in this work is the temperature profile. The polymer in the region of the die will be roughly at the set temperature for the die and billet, assuming isothermal conditions in this region. As the polymer necks away from the die and the product moves axially away from the die it will cool progressively. It was observed that drawing down occurred not only in the die region, but also outside of the die region, i.e. the higher draw ratio product continued to draw for some distance from the die (*Figure 11*). The extent of this post-die drawing zone will be related to the material, draw speed and set drawing temperature and would be expected to increase with draw speed and set drawing temperature. As yet, we do not have sufficient quantitative information to describe this zone in detail. It is clear, however, that drawing will effectively cease when the stress at the highest deformation ratio element becomes insufficient to deform the material at the prevailing temperature of that element, i.e. a stable product is obtained. When drawing is terminated there will be two sections of the product which have not attained the desired $\lambda_{max}$, namely the section corresponding to the die and post-die deformation zones, and also the section near to the drawing clamp. For a fixed set of drawing conditions, increasing the product length will decrease the proportion of the total length occupied by these 'waste' zones, and a continuous process would render them of insignificant length compared to the desired product length. Clearly, quenching of the product would also be a method of controlling the value of $\lambda_{max}$ by reducing or eliminating the post-die deformation; alternatively, post-die heating may prove to be desirable in certain cases.

The results shown in *Figure 9* indicate that a desired $\lambda_{max}$ could be obtained by choice of a suitable $R_N$ and draw speed. As yet the effect of nominal temperature on the process has not been investigated, but a family of curves

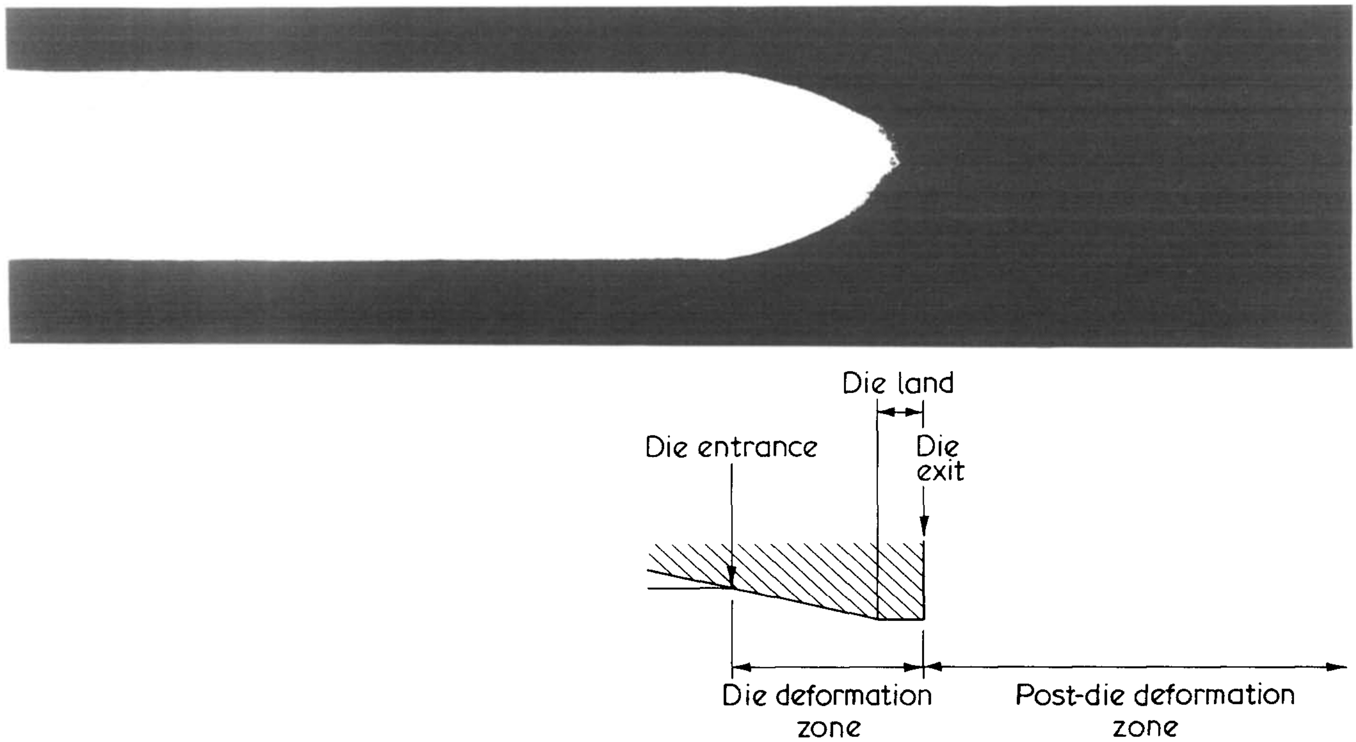


*Figure 11* PP copolymer $R_N$ = 7 billet removed from die showing the die deformation zone and part of the post-die deformation zone.

similar to those presented in *Figure 9* would allow specification of drawing conditions for obtaining a desired $\lambda_{max}$ with similar accuracy to that attainable in hydrostatic extrusion.

As the draw speed is increased and higher draw ratios are produced it is possible that the temperature gradient along the product may be altered considerably: deformation in the die region, and possibly in the part of the post-die zone may tend towards an adiabatic state, where the work done in deformation is not dissipated quickly to the surroundings. This effect might be expected to be more pronounced in the larger scale drawing process. It is difficult to identify such trends clearly in the present work, although the stress values for the small scale process do tend to lie slightly higher than those for the large scale process (*Figure 8*) especially at $R_N$ = 5 at the higher draw speeds. Truly isothermal data (e.g. obtained from a temperature-controlled environment) are required for assessment of any adiabatic effects. In the limit, a totally adiabatic process could lead to an unstable process with probable fracture in the die region, i.e. the neck may not be stabilized. Further investigations into the stability and control of die drawing are being undertaken at present.

*Other materials.* Certain polymers which are capable of being deformed to high draw ratios, e.g. on a tensile testing machine, prove to be extremely difficult to extrude hydrostatically either to high deformation ratios or, at practical rates, due to large effects of pressure on their deformation, and/or the adverse strain rate field discussed below.

Die drawing offers the considerable advantage of being a method–possibly the only method– of obtaining large section rods of these materials for possible engineering use. One notable example is die drawing of POM to a draw ratio of ~ 12.5 with considerable ease[16] whereas deformation ratios of ~ 10 become quite impractical in hydrostatic extrusion of POM[9]. Also being investigated at present are various grades of LPE[20], and different product cross-sections, and it is intended to present the results of these investigations in separate publications. A particular requirement for materials, especially for large scale die drawing (or extrusion), is that the necessary billets can be successfully manufactured if they cannot be purchased.

*Die Drawing Products*

Die drawing has been found to be a controllable method of obtaining high draw ratio rods exhibiting greatly enhanced values of axial Young's modulus, *E*, up to ~ 20GPa. Comparisons of die drawing products with fibre drawing and extrusion (*Figure 10*) indicate that a similar change in stiffness with draw ratio is being obtained in each process, i.e. they all appear to be providing a similar degree of effective deformation. It is worth noting here that the fibre drawing and extrusion results refer to PP homopolymers, whereas the present work concerns a PP copolymer of lower initial stiffness than the homopolymer. In considering the effectiveness of the deformation, it is salient to recall the temperature gradient involved in our die drawing experiments. If the temperature is considered to fall with distance from the die, then the higher draw ratio element of the product will be drawn at progressively decreasingly temperatures, i.e. under conditions which may tend to favour more effective drawing[4].

It is not clear from *Figure 10* if significant deformational heating has occurred, as discussed above. Deformational heating would lead to an increase in the actual drawing temperature and so might be expected to reduce the stiffness obtained for a given $\lambda_{max}$[3]. The decrease in slope of the modulus–draw ratio curve at high draw ratios is common to the three modes of deformation represented in *Figure 10*, and of these, hydrostatic extrusion will probably represent the nearest to the ideal isothermal situation. The decrease in slope may therefore not be indicative of significant deformational heating, but rather may be a feature of the mechanical properties of highly deformed PP. However, the decrease in slope of the $E-\lambda_{max}$ curve appears to be greatest for die drawing, which may reflect a measure of deformational heating at the highest deformation ratios.

A feature of the appearance of the die drawn products is the general increase in transparency with deformation, typical of hydrostatically-extruded rods, but not always observed in tensile tests. This transparency persists up to $\lambda \sim 17.1$, but the highest draw ratio specimen is quite opaque. The transparency at lower draw ratios may be aided by the polymer being in contact with the die wall in the early stages of deformation: the compressive stress normal to the die wall may help to eliminate voiding.

It should be noted that the highest draw ratio reported in the present work does not necessarily represent the limit for die drawing PP; it is to be expected that higher draw ratios will be attainable at higher drawing speeds, and it will be interesting to observe if these are effective in further increasing the stiffness.

## CONCLUSIONS

(1) Die drawing has been found to be a controllable process for the production of very high stiffness PP rods, and has the following advantages:

(i) the higher the desired draw ratio, the greater is the production rate at a given nominal temperature;

(ii) there are no significantly adverse pressure effects, unlike hydrostatic extrusion, and the strain rate field in the deformation region is not imposed totally by the geometry of the die;

(iii) the required equipment is relatively simple (compared to, say, extrusion equipment);

(iv) the process has the potential for development into a continuous process, or may be operated as a batch process if required.

Consequently the process should have powerful application to those materials which can be highly drawn, but for which high deformation ratios cannot be attained in hydrostatic or ram extrusion, as well as to those materials in which high extrusion ratios can be attained. The process clearly may also have application, especially for large sections, to materials which cannot be highly drawn.

(2) Very high stiffness PP copolymer rods (up to 20.6 GPa at $\lambda = 19.9$) have been successfully produced, several metres long. These rods were all unflawed (except for the highest draw ratio being opaque). The increase in axial Young's modulus with draw ratio was very similar to that observed in fibre drawing and hydrostatic extrusion of PP homopolymer.

## NOTE:

The work described in this paper and similar work on other polymers is the subject of a UK Patent Application (No. 7919737, filed 6 June 1979)

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## REFERENCES


1 Capaccio, G., Crompton, T. A. and Ward, I. M. *J. Polym. Sci. (Polym. Phys. Edn)* 1976, **14**, 1641
2 Brew, B. and Ward, I. M. *Polymer*, 1978, **19**, 1338
3 Cansfield, D. L. M., Capaccio, G. and Ward, I. M. *Polym. Eng. Sci.* 1976, **16**, 721
4 Coates, P. D. and Ward, I. M. *J. Mater. Sci.* 1978, **13**, 1957
5 Wilding, M. A. and Ward, I. M. *Polymer*, 1978, **19**, 969
6 Gibson, A. G. and Ward, I. M. UK Pat. Appl. 30823/73 (filed June 1973)
7 Gibson, A. G., Ward, I. M., Cole, B. N. and Parsons, B. *J. Mater. Sci.* 1974, **9**, 1193
8 Gibson, A. G. and Ward, I. M. *J. Polym. Sci. (Polym. Phys. Edn.)* 1978, **16**, 2015
9 Coates, P. D. and Ward, I. M. *J. Polym. Sci. (Polym. Phys. Edn.)* 1978, **16**, 2031
10 Williams, T. *J. Mater Sci.* 1973, **8**, 59
11 Hope, P. S. and Parsons, B. to be published
12 Coates, P. D. Gibson, A. G. and Ward, I. M. *J. Mater. Sci.* in press
13 Bunney, J. B. and Cassin, C. (ICI), UK Pat. No. 1311885
14 Coates, P. D. and Ward, I. M. to be published
15 'Ultra-High Modulus Polymers', (Ed. A. Ciferri and I. M. Ward), Applied Science Publishers, London, 1979, Chapters 1 and 10
16 Coates, P. D. unpublished work
17 Gibson, A. G., Davies, G. R. and Ward, I. M. *Polymer*, 1978, **19**, 683
18 ICI Technical Service Notes PP100 and PP TD527
19 Parnaby, J., Battye, P. G., Hassan, G. A. and Hadwell, C. P. *Plastics & Rubber; Processing*, **89**, Sept. 1978
20 Gibson, A. G., Coates, P. D. and Ward, I. M. *J. Mater. Sci.*, in press