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Auxetic polyproplyene fibres have been produced on a large-scale industrial extruder for the first time. A first batch of tests identified a coarse processing window which was then more closely defined by subsequent tests. The effects of barrel temperature, screw speed, take-up speed and quenching (through the parameters of air gap and bath temperature) on the Poisson's ratio and Young's modulus of the fibres were examined. It was found that a temperature of 200ºC, screw speed of 12.5rpm and take-up speed of either 1.5 or 3.5rpm produced fibres with a high degree of auxeticity up to 5% strain – significantly increasing the auxetic strain range previously reported for lab-scale extrusions at lower extruder temperatures. Quenching with a water bath reduced the auxeticity, so was not employed as a processing parameter.

1 Introduction Auxetic materials [1] are those which have a negative Poisson's ratio, ν. Poisson's ratio is defined as the negative ratio of the transverse strain to longitudinal strain for a uniaxial mechanical stress acting in the longitudinal direction. This means that when auxetic materials are stretched in the longitudinal direction, they expand laterally rather than contract as a conventional material would do. This counterintuitive property is found in natural materials such as certain forms of skin [2] and bone [3] and minerals such as α-cristobalite [4]. As well as identifying and understanding these natural auxetic materials, much of the recent work has focussed on engineering auxetic behaviour into synthetic materials by dint of their microstructure. The first reported synthetic auxetic material was a polymeric foam, developed by Lakes in 1987 [5]. In 1989, Evans and Caddock examined a commercially available form of expanded polytetrafluoroethylene (PTFE) [6, 7] and found that it possessed a large negative Poisson's ratio of up to -12 after pre-conditioning. For isotropic materials the thermodynamically allowed lower limit on Poisson's ratio is -1 and so the value reported in PTFE is extremely low and is only possible in strongly anisotropic materials. The causal mechanism for this was based on its complex microstructure, which consisted of nodules interconnected by fibrils. Depending on the connectivity of such microstructures, deformation can predominantly be either by rotation of the fibrils (or ligaments) [7] or rotation of the nodules [7, 8] in response to a mechanical load. In the case of microporous PTFE, fibril rotation ('hinging') causing the nodules to translate was shown to describe the auxetic effect observed experimentally at low and intermediate tensile strain very well [7]. A secondary simultaneous mechanism of fibril stretching in the same microstructural connectivity accounted for the transition to positive Poisson's ratio at high tensile strain [9]. This combined mechanism was also found to describe the full strain-dependent Young's modulus behaviour very well. Nodule rotation was additionally shown to lead to a reduction in magnitude of negative Poisson's ratio at higher strain [7] – but this required an alternative connectivity to the low/intermediate strain microstructure and did not produce the observed positive Poisson's ratio at high strain.

There appeared at this stage to be no reason to suspect that PTFE was unique in its ability to be processed to produce this microstructure, so attempts were made to process

other polymers into an auxetic form. This was successfully achieved for ultra-high molecular weight polyethylene (UHMWPE) [10], polypropylene (PP) [11] and nylon [12]. In all cases, the processing route used conventional partial sintering to achieve surface melting of the particles only [13] followed by extrusion [14], with the entire process taking place within the barrel of a bench-top extruder. To ensure structural integrity, a compaction stage was used [15] prior to the sintering stage. The polymers were produced in the form of cylindrical rods of diameter 9-15mm, depending on the diameter of the extruder barrel, and length 100mm.

In 2002, building on the work on extruded cylinders, a process was reported [16] which produced auxetic polymeric fibres. The initial work was carried out using PP as this was a material which was well understood for the processing of auxetic polymers. For example, PP was used to assess the effect of powder morphology [11] on the auxeticity of the final polymer. To this end, the same grade of polymer was used as had been successfully employed in the cylinder work i.e. PB0580 produced by Plast-Labor S.A. The fibres were found to have a granular, or fused-particle, microstructure [16, 17], with little evidence of the nodule-fibril microstructure reported for the earlier cylindrical rods. The process was carried out on an Emerson and Renwick Ltd. Labline extruder which had a 3:1 compression ratio, and a screw of diameter 25.4mm and length/diameter ratio 24:1. A particular feature of this extruder is that it has 5 thermostatically controlled temperature zones. However, in order to produce auxetic fibres, it was necessary to use a temperature of 159ºC in each of the zones.

A detailed investigation of the processing parameters [17] defined conditions of screw speed 10rpm (equating to $1.05 rads^{-1}$) and a take-up speed of $0.032 ms^{-1}$. The fibres produced had moduli of around 1.3GPa and an average Poisson's ratio, ν = -0.6, with the most auxetic sections having ν = -1.62. Drawing after extrusion to improve the modulus was not an option in this process because it was found that this reduced auxeticity. Initially, the die used consisted of a 40-hole spinneret plate with each hole of diameter 0.55mm, producing fibres of diameter ~ 340μm. Interestingly, when this spinneret plate was replaced by a monofilament plate, with a 1mm diameter hole, this also produced fibres of diameter ~ 340μm, but the fibres were more consistently auxetic [17]. The process was also adapted to produce auxetic polyester and nylon fibres [17] and also auxetic PP films [18]. In the case of auxetic PP films, auxetic behaviour has been produced for extrusion at 159ºC [18] and also at elevated extrusion temperature around 180-190ºC [19].

An alternative to the extruded monofilament approach to making auxetic fibres is the helical auxetic yarn (HAY) consisting of a thin stiff yarn wrapped helically around a thick compliant core yarn [20]. As the HAY is stretched lengthwise the thin wrap yarn straightens and pushes the core yarn into a helical configuration, producing an overall increase in the thickness of the composite yarn system and, therefore, auxetic response. This approach requires careful control of the wrap yarn placement on the core yarn, and it has been shown that the magnitude and presence of the auxetic effect is dependent on wrap yarn angle and the core and wrap yarn diameters and moduli [21, 22]. A variant on the HAY is to embed the thin stiff component within the thick compliant core [23].

Auxetic fibres have a wide range of potential applications, both making use of the auxetic effect itself and of the property enhancements due to the negative Poisson's ratio. As an example of the former, auxetic fibres within a composite have been shown to resist the failure mechanism of fibre pullout [24,25]. Effectively here, as the fibre is pulled, it expands, causing it to lock into the matrix rather than contracting and pulling away from the matrix as in the conventional case. Looking now at the properties of auxetic materials, many of these are predicted to be enhanced due to the presence of a negative Poisson's ratio. These properties include fracture toughness, wear resistance and energy absorption [26, 27], all of which could be extremely useful in textiles for, say, personnel protective applications.

One of the limiting factors in the move from laboratory production to commercialisation of extruded auxetic monofilament fibres is the fact that they are currently produced on a laboratory extruder, albeit a semi-commercial one. This paper examines the preliminary stages in moving to a large scale extruder and the effect of this on the key processing parameters.

2 Experimental methods

2.1 Powder characteristics The powder used in these extrusions was PP4352F1-50962 BGNRI 09430/31/33/34/36/39/42, produced by Exxon Mobil Chemical. This was then Messier ground to 92.5% of particles less than 500μm diameter; 32.0% of particles less than 250μm diameter. The key processing temperatures were measured using differential scanning calorimetry (DSC) at a heating rate of 10ºC/min under a flow of nitrogen at $20ml\ min^{-1}$. These revealed that the melting temperature was 165.7ºC, with the onset of melting temperature being 142ºC.

2.2 Fibre extrusion A schematic of a general melt extruder is shown in Fig. 1a. In conventional melt extrusion polymer granules or powder are fed into the hopper and transported through the heated barrel of the extruder by the screw. The molten polymer passes through a metering pump which regulates the throughput rate, prior to entering the die zone. The polymer passes through a spinneret plate comprising of a series of holes which impart cross sectional shape to the extruded fibres as the polymer passes through the spinneret. On exiting the spinneret the fibres undergo rapid cooling in an air quench zone, either under ambient air conditions (ambient air quench) or using an air

knife, and may also enter a quench tank containing cooling water. The quenching process facilitates the solidification of the extruded fibres by cooling them below the polymer melt crystallization temperature and eventually the glass transition temperature. After quenching, the fibres enter into a system of take-up, draw and final winding rollers. The take-up winder speed, together with the throughput rate from the metering pump, determines the denier (linear mass density) of the fibre. Draw rollers can impart additional drawing of the fibres which, in conventional melt extrusion, aligns polymer chains to impart increased strength and stiffness.

The extruder used for these tests was a Davis-Standard Thermatic extruder based at Shakespeare Monofilament UK Ltd, as shown in Fig. 1b This was a much larger machine than that previously used, but still had 5 electrically heated barrel zones with individual thermostatic controls. The screw was driven by a 50 hp DC motor and had a screw length of 1450mm and screw diameter of 50mm, giving a length/diameter ratio of 29:1. To produce the fibres, the extruder was fitted with a spinneret plate, which in this case had 20 die slots, each of diameter 2mm. The extrusion line also had the facility for both air and water quenching (Fig. 1c) as the fibres exit the die slots.

The first batch of extrusions was conducted to try to identify the boundaries of the processing window and was thus relatively coarse. The key processing parameter for production of auxetic polymers has already been stated to be the extrusion temperature. For PP, this has been 159ºC for cylinder and fibre production using the PB0580 powder in small bore barrel batch (10-15mm diameter) [11] and continuous (25.4mm diameter) [17] extruders, respectively. Auxetic PP films have been produced from the same powder on the 25.4mm diameter extruder at 159°C and also a higher temperature of 180-190ºC [19]. Attempts to run the larger diameter extruder of the current investigation at 159ºC proved to be difficult due to very high pressure build-up in the die region. So, on the advice of the machine Technicians, the temperature was raised so that safe extrusion could proceed.

The set of related temperatures, screw speeds and take-up speeds used in the first batch of tests are listed in Table 1. Conditions were defined based on previous studies using the experience of manufacturing auxetic PP cylinders, fibres and films and the complex interaction known to exist between temperature, screw speed and take-up speed. This allowed the investigators to define a set of conditions taking into account extruder variables. In total, 14 sets of conditions were examined, with temperatures varying from 180 – 210ºC, screw speeds from 12.5 to 18.8rpm and take-up speeds from 1.5 – 3.5 rpm. A new variable was also investigated which was not possible with the laboratory scale runs, i.e. the effects of quenching. The fibres were either subjected to ambient air quench as they exited the die or were water quenched at 21 or 30ºC. At this stage, the air gap to the water bath was not varied.

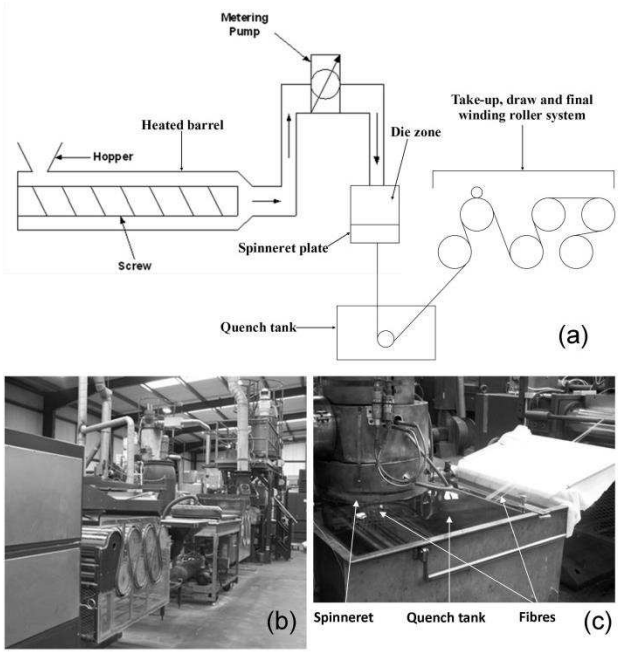

Figure 1 (a) Schematic of a general melt extruder. (b) The Davis-Standard Thermatic Extruder used in the large scale extrusion; (c) Spinneret and quench tank of the Davis-Standard Thermatic Extruder.

Table 1 Processing parameters for the first set of tests.

Fibre number	Temperature (ºC)	Screw speed (rpm)	Take-up speed (rpm)	Quench temperature(ºC)
PP1	180	18.8	1.5	30
PP2	180	12.5	1.5	30
PP3	180	12.5	1.5	21
PP4	182	12.5	1.5	21
PP5	200	12.5	1.5	Ambient
PP6	200	12.5	1.5	21
PP8	200	18.8	1.5	21
PP9	200	12.5	3.5	30
PP10	200	12.5	3.5	Ambient
PP11	200	12.5	1.5	Ambient
PP12	200	12.5	3.5	Ambient
PP13	207	12.5	3.5	30
PP14	210	12.5	3.5	30

After analysis of the fibres produced, a second set of extrusions were undertaken. This was to examine more closely the processing parameters and their effect on auxeticity. The first batch of tests had identified the optimum temperature to use and this was employed for all the subsequent runs. The first run of the second batch of extrusions fixed the temperature and the take-up speed and allowed the screw speed to vary as 12.5, 14.5, 15.5, 16.5 and 18.5rpm (see Table 2). The second run (see Table 3) fixed the temperature and screw speed and allowed the take-up speed to vary as 1.5, 2.5, 3.5, 4.5 and 5.5 rpm. For both cases, ambient air quenching was carried out.

Table 2 Processing parameters used to investigate the effects of screw speed, with the other parameters fixed.

Fibre number	Temperature (ºC)	Screw speed (rpm)	Take-up speed (rpm)	Quench temperature (ºC)	Air Gap (mm)
PP10	200	12.5	3.5	Ambient	-
PP12	200	12.5	3.5	Ambient	-
PP15	200	12.5	3.5	Ambient	-
PP17	200	14.5	3.5	Ambient	-
PP18	200	15.5	3.5	Ambient	-
PP19	200	16.5	3.5	Ambient	-
PP20	200	18.5	3.5	Ambient	-

Table 3 Processing parameters used to investigate the effects of take-up speed, with the other parameters fixed.

Fibre number	Temperature (ºC)	Screw speed (rpm)	Take-up speed (rpm)	Quench temperature (ºC)	Air Gap (mm)
PP5	200	12.5	1.5	Ambient	-
PP11	200	12.5	1.5	Ambient	-
PP16	200	12.5	1.5	Ambient	-
PP27	200	12.5	1.5	Ambient	-
PP24	200	12.5	2.5	Ambient	-
PP10	200	12.5	3.5	Ambient	-
PP12	200	12.5	3.5	Ambient	-
PP15	200	12.5	3.5	Ambient	-
PP25	200	12.5	4.5	Ambient	-
PP26	200	12.5	5.5	Ambient	-

Tables 4 and 5 give the details for the third and fourth runs, which concentrated on quenching. The third run (Table 4) fixed the temperature, screw speed and take-up speed along with the use of a 20ºC quench. The air gaps varied from 20 to 40 and 60mm and the fibres were compared with fibres produced using the same processing parameters under ambient air quenching. The final run of tests (Table 5) used a fixed temperature, screw speed and take-up speed. An air gap of 60mm was employed and the quench temperature was varied from ambient air quench to 20, 30, 40, 50, 60 and 70ºC.

2.3 Fibre characterisation The fibres were tested using an Instron 4300 mechanical testing machine, fitted with a 100N load cell, at a crosshead speed of 2mm min^{-1}. The Poisson's ratio was measured using a Messphysik ME46 videoextensometer. This consists of a computer software package, which simultaneously measures length and width data from changes in the contrast between markers attached to the fibres along their lengths and the edges of the fibres, which gave the width measurement. The schematic set up of the equipment is shown in Figure 2. The software splits the length defined by the fiducial markers into 10 segments, the widths of which are then tracked throughout the test. Occasionally, individual segments can be tracked incorrectly due to features on the fibre surface, lighting issues and such like. This is evident through the recorded segment width being significantly different to the actual fibre diameter (i.e. adjacent segment widths), or displaying markedly different trends to other segments. In such cases, the identified segment data were removed from the subsequent analysis.

Table 4 Processing parameters used to investigate the effects of the air gap, with the other parameters fixed.

Fibre number	Temperature (ºC)	Screw speed (rpm)	Take-up speed (rpm)	Quench temperature (ºC)	Air Gap (mm)
PP10	200	12.5	3.5	Ambient	-
PP12	200	12.5	3.5	Ambient	-
PP15	200	12.5	3.5	Ambient	-
PP21	200	12.5	3.5	20	20
PP22	200	12.5	3.5	20	40
PP23	200	12.5	3.5	20	60

Table 5 Processing parameters used to investigate the effects of the quench temperature, with the other parameters fixed.

Fibre number	Temperature (ºC)	Screw speed (rpm)	Take-up speed (rpm)	Quench temperature (ºC)	Air Gap (mm)
PP5	200	12.5	1.5	Ambient	-
PP11	200	12.5	1.5	Ambient	-
PP16	200	12.5	1.5	Ambient	-
PP27	200	12.5	1.5	Ambient	-
PP28	200	12.5	1.5	20	60
PP34	200	12.5	1.5	20	60
PP29	200	12.5	1.5	30	60
PP30	200	12.5	1.5	40	60
PP31	200	12.5	1.5	50	60
PP32	200	12.5	1.5	60	60
PP33	200	12.5	1.5	70	60

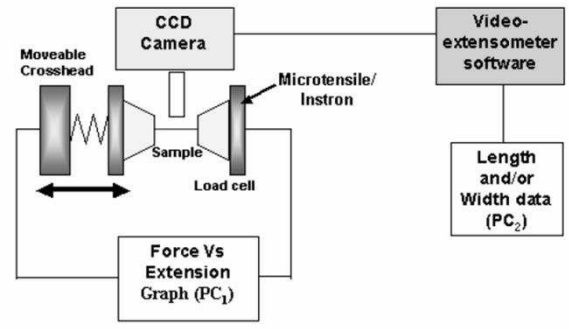

Figure 2 Schematic of the videoextensometry set up.

3 Results and Discussion The modulus, diameter and Poisson's ratio for the initial batch of tests are given in Table 6. The data are averages from several tests, and for

Poisson's ratio are averaged over all width segments (following removal of any outliers – section 2.3) for several tests. As an example, Fig. 3a shows the raw data for PP14. This clearly shows as the length increases (bold line), the width decreases. This is conventional behaviour and is found in all sections of the fibre. To get the Poisson's ratio from these data, an average true transverse strain vs true axial strain plot is constructed, and the slope of this gives -ν. In this case, ν = +0.36 (Fig. 3b). Figure 4 shows the same analysis for PP13, which is an auxetic fibre. It can be seen clearly that as the fibre is pulled, it increases in diameter (Fig. 4a), giving the Poisson's ratio by analysis as above as ν =-0.82 (Fig. 4b). The auxetic effect is then shown for the first time in a fibre produced on a large-scale extruder, and persists over the full 5% axial strain range covered in the test. This is an increase in the strain range for auxetic behaviour found in melt extruded auxetic fibres which persists typically up to 1-2% strain in fibres produced previously on a lab-scale extruder.

Table 6 Mechanical properties for the first batch of tests.

Fibre number	Diameter (mm)	Young's modulus (GPa)	Poisson's ratio
PP2	2.02	0.65±0.02	0.00±0.03
PP3	2.08	0.79±0.02	0.32±0.04
PP4	2.27	0.68±0.01	-0.41±0.05
PP5	1.82	1.05±0.03	-0.47±0.05
PP6	2.03	1.04±0.03	-0.24±0.03
PP8	1.66	0.96±0.07	0.35±0.05
PP9	1.72	0.87±0.05	0.35±0.04
PP12	1.86	1.08±0.04	-1.0±0.1
PP13	1.30	1.06±0.04	-0.8±0.1
PP14	1.60	0.91±0.05	0.31±0.05

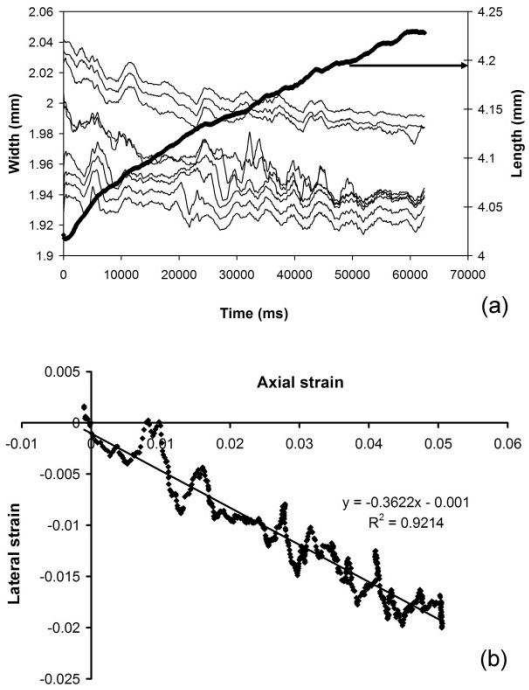

Figure 3 (a) Raw width and length data for PP14, showing conventional behaviour; (b) Plot of lateral strain against axial strain for PP14, showing Poisson's ratio, ν = +0.36.

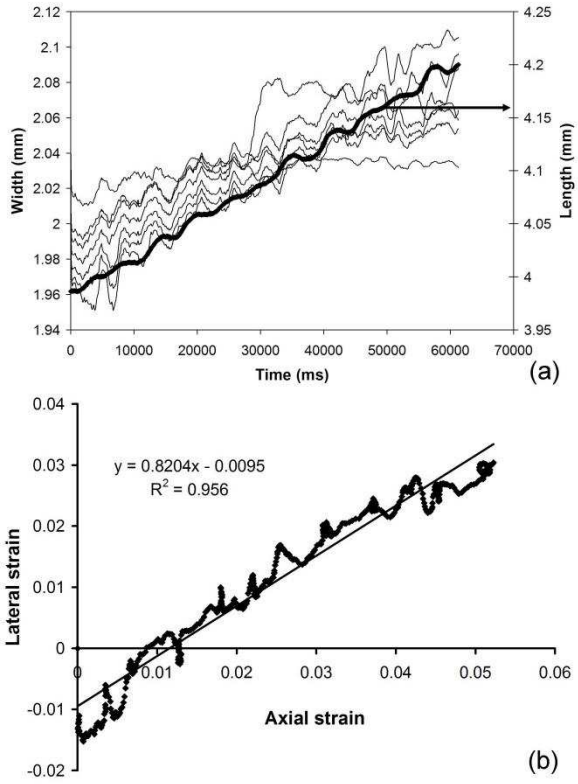

Figure 4 (a) Raw width and length data for PP13, showing auxetic behaviour; (b) Plot of lateral strain against axial strain for PP13, showing Poisson's ratio, ν = -0.82.

The results shown in Table 6 appear to indicate that the best temperature to use for the detailed processing parameter examinations is 200ºC. Tests conducted at 180ºC produced near zero or positive Poisson's ratio fibres and were produced at the machine limits due to the pressure build-up described above. It is noted that PP4 was auxetic and was produced at 182ºC, but it was felt that this temperature was very close to 180ºC and machine fluctuations might result in conventional fibres being produced. PP14, produced at 210ºC, was also conventional, so 200ºC was selected as the temperature. Table 6 appears to indicate that increasing the screw speed or take-up speed results in the fibre being conventional even at a processing temperature of 200ºC. This is investigated further below.

In the analysis that follows, the data are plotted as a range of Poisson's ratio values (each averaged over all width segments during one test) for several tests at each condition, rather than as a single value. The latter is useful for a coarse examination as was undertaken above. However, in the majority of work carried out on extruded fibres, plotting all the data as a range is the usual way of representing the data allowing the recognised variability of the fibres to be shown, see for example [17]. This also allows the auxeticity of the fibres to be clearly displayed, meaning that one very negative (or indeed positive) value did not skew the results.

3.1 Effect of screw speed Figure 5 shows the effect of screw speed on the Poisson's ratio range (a) and the Young's modulus (b) of the PP fibres for the tests conducted according to Table 2. The temperature was fixed at 200ºC and a take-up speed of 3.5rpm was used. At a screw speed of 12.5rpm, all the fibres tested were auxetic but had a low modulus. Increasing the screw speed results in some fibres with auxetic character, but these were predominantly conventional. This is in agreement with previous findings on cylinders and fibres where in order to achieve an auxetic polymer by extrusion, either in a benchtop extruder or in a lab-scale melt extruder, there needs to be a defined dwell time to allow the polymer to surface melt. It is also interesting to see that the most auxetic fibre has the lowest modulus, again as has previously been observed.

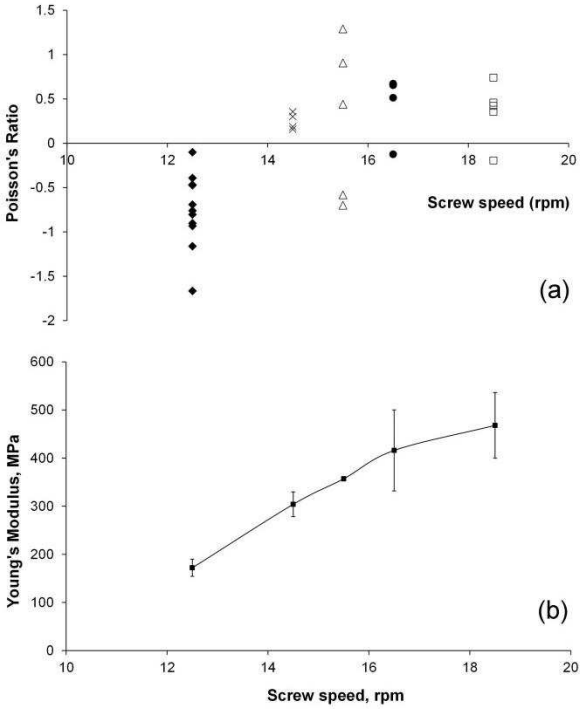

Figure 5 (a) Effect of the screw speed on the Poisson's ratio values of the fibres; (b) Effect of the screw speed on the Young's modulus of the fibres. Temperature = 200°C and take-up speed = 3.5rpm; ambient air quenching.

For the remaining tests, then, the temperature remained at 200ºC and the screw speed was 12.5rpm.

3.2 Effect of take-up speed Figure 6 shows the effect of take-up speed on the Poisson's ratio range (a) and the Young's modulus (b) of the PP fibres for the tests conducted according to Table 3. Auxetic character is shown for all the take-up speeds investigated, with only the faster speed of 5.5rpm showing any large positive Poisson's ratios. At a take-up speed of 1.5rpm, all but 1 test conducted showed auxetic behaviour and at 3.5rpm, all the tests showed auxetic behaviour. So, either of these two can be used to produce auxetic fibres. Figure 6b shows that the fibre modulus decreases as the take-up speed increases. This is exactly the same finding as was seen with the laboratory scale fibres [17] and was attributed in that case to slight drawing of the fibre structure, which again can be assumed here.

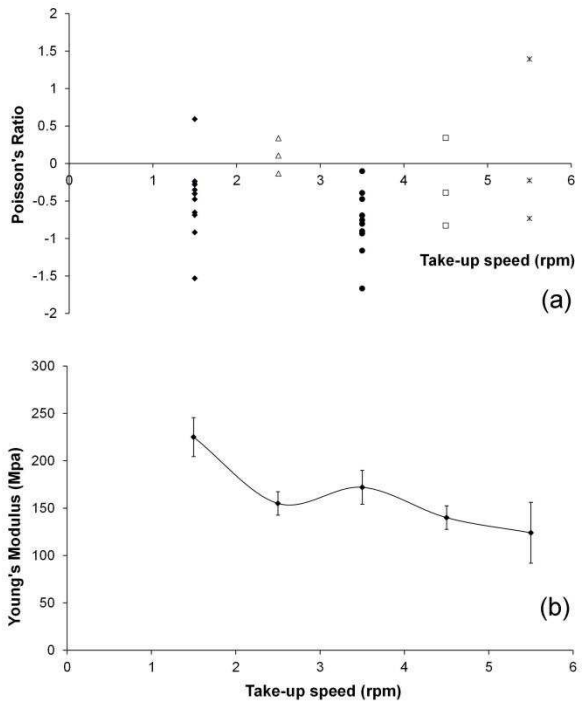

Figure 6 (a) Effect of the take-up speed on the Poisson's ratio values of the fibres. Each take-up speed is represented by the full range of the data points obtained; (b) Effect of the take-up speed on the Young's modulus of the fibres. Temperature = 200°C and screw speed = 12.5rpm; ambient air quenching.

3.3 Effect of varying the air gap to the quench bath Figure 7 shows the effect of varying the air gap to the quench bath on the Poisson's ratio (a) and the Young's modulus (b) for the PP fibres for test conditions according to Table 4. In this case, the Young's modulus of the fibres decreased as the air gap increased. So, given that the air gap appeared to have very little effect on the Poisson's ratio with only the 40mm gap showing any positive character and this not being the case with the 60mm air gap, then for the next set of tests investigating the quench temperature, the easiest distance to work with (60mm) was used.

3.4 Effect of quench bath temperature Figure 8 shows the effect of varying the quench bath temperature on the Poisson's ratio (a) and the modulus (b) for the fibres produced according to the parameters in Table 5. The first thing to note is that quenching with the water bath appears to reduce auxeticity (Fig. 8a). With ambient air quench, all the fibres were auxetic, with values down to $\nu = -1.5$. Taken together with Fig. 8b, which shows that any amount of water bath quenching also reduces the modulus, it appears that the best condition would be to ambient air quench the fibres.

The quenching data suggest that the required combination of microstructure and mechanism responsible for auxetic behaviour in part evolves after exiting the die. Quenching at the lowest temperatures of 20-30°C shows no evidence of auxetic character. It may be at these temperatures the water quench may not allow the thermal rearrangement or evolution of the molecular chains and/or granular structure into the configuration required for auxetic response. Increasing the quenching temperature may, on the other hand, allow some thermal rearrangement/evolution of the microstructure-mechanism such that there is an increased tendency for auxetic behaviour at the highest quenching temperature of 70°C. That the ambient air quench does produce auxetic behaviour, whereas the lowest water quench temperatures in the vicinity of the ambient air temperature do not, may be due to the less efficient heat transfer in air than in water allowing increased time for the necessary microstructural rearrangements to occur.

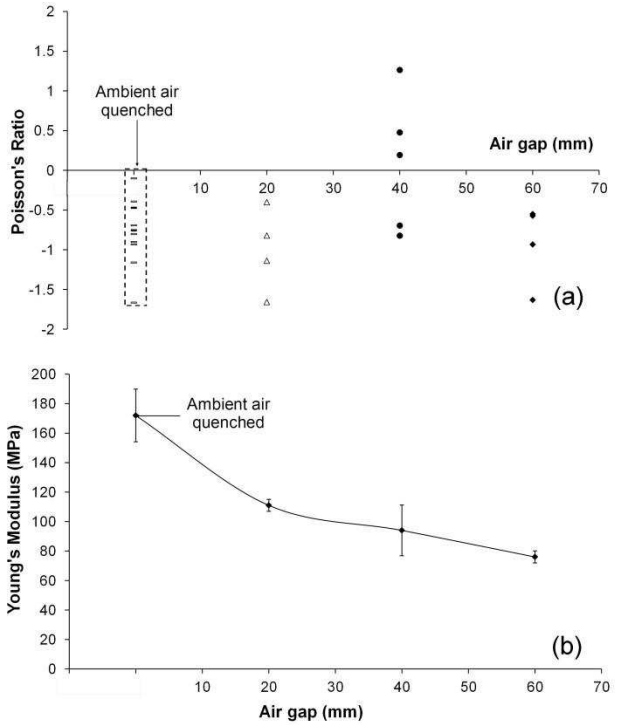

Figure 7 (a) Effect of the air gap on the Poisson's ratio values of the fibres. Each air gap in mm is represented by the full range of the data points obtained; (b) Effect of the air gap on the Young's modulus of the fibres. Temperature = 200°C, screw speed = 12.5rpm, take-up speed = 3.5rpm and quench temperature = 20°C.

3.5 Reproducibility Taking all the tests into consideration, the following conditions can be defined from this preliminary study to produce auxetic behaviour. These are a barrel temperature of 200ºC, a screw speed of 12.5rpm, a take-up speed of either 1.5 or 3.5 rpm and ambient air quench.

To assess the reproducibility when scaling up the process, the two sets of conditions which are defined as optimum are compared in Fig. 9. It can be seen that reproducibility is good for a take-up speed of 1.5rpm but even better for a take-up speed of 3.5rpm, where all the fibres produced were auxetic.

At this stage, it is worth considering the challenges which occur in actually carrying out the extrusion in an industrial scale extruder and which need to be addressed to fully commercialise the process. The hopper feed became blocked frequently, leading to issues in feeding powder to the screw. The fibres were difficult to feed to the spooler, causing broken fibres and tangles, which had also been a problem on the laboratory scale extruder. Finally, running the extruder at lower temperatures caused instability in the temperatures, leading to a lack of necessary control which meant that some of the runs had to be aborted. It is likely that these processing and control issues are at least in part responsible for the significant variability evident in the Poisson's ratio and Young's modulus data in Figs. 5-9. All of these issues need to be addressed in order to optimise fully the process for the production of fibres showing reduced variability in properties.

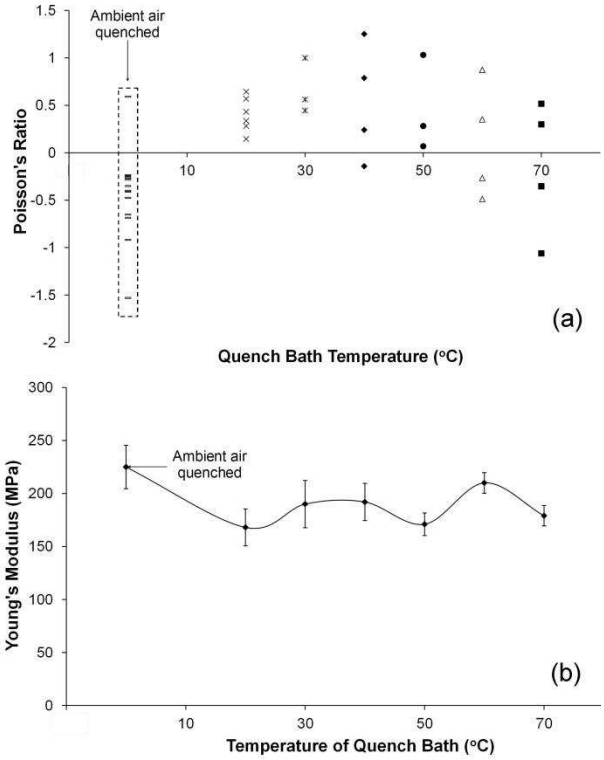

Figure 8 (a) Effect of the quench temperature on the Poisson's ratio values of the fibres. Each quench bath temperature is represented by the full range of the data points obtained; (b) Effect of the quench temperature on the Young's modulus of the fibres. Temperature = 200°C, screw speed = 12.5rpm, take-up speed = 1.5rpm and quench gap = 60mm.

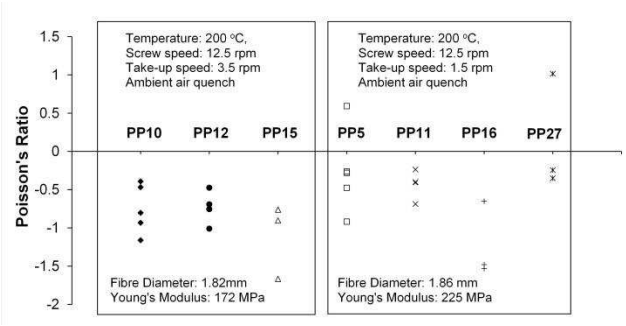

Figure 9 Comparison of the data for the defined processing conditions to illustrate the reproducibility of the fibres.

The most striking of the processing parameter variables studied here in terms of the results gained was the temperature. It was expected that, in common with the previous work on auxetic PP fibres [17] and cylinders [11] to date, the temperature would need to be around 159ºC, give or take a few degrees adjustment for the change in powder. The barrel temperature of 200ºC identified in this work for the production of auxetic fibres when scaling up the process to industrial-size extruders was, therefore, unexpected. It is possible that the higher barrel temperature in the large-scale extruder reproduces similar conditions in the bulk polymer during the extrusion process to the lower temperature in the smaller scale extruder reported previously – thought to lead to a granular microstructure responsible for the auxetic effect in the fibre [16, 17], through fusing of surface-melted powder particles. Alternatively, the large scale extrusion temperature may produce conditions in the bulk polymer similar to those recently discovered to lead to auxetic behaviour in PP films in the smaller scale extrusion process at 180ºC [19]. In this latter case, the polymer is likely to be fully molten during extrusion and so an alternative fibre microstructure for auxetic behaviour is likely.

The thermal characteristics of the polymer in the large scale extruder, and the resulting fibre microstructure, are not known and this is the main focus of future work. The focus will be to determine the physical state of the polymer in the larger barrel during extrusion at 200ºC. This will be related to the corresponding state in the smaller barrel laboratory extruder at the 159ºC conditions established for auxetic fibres, and also the additional 180-190ºC conditions established for auxetic PP films. It will be necessary to establish the effects of dwell time and thermal equilibration on the nature of the polymer in situ (e.g. homogeneous melt or a partially melted, heterogeneous state). A post-production assessment of fibre morphology and crystallinity will also be undertaken to determine the microstructural features and size scale responsible for the auxetic effect. This detailed investigation will not only enable a detailed understanding of the processes in the small and larger scale extruders used to date, but will also facilitate transfer of the process to a broader range of other extruders having different screw dimensions.

The lower limit on the diameter of fibres produced at 159ºC in the smaller extruder is likely to be determined by the size of granular (fused-particle) features making up the fibre microstructure. The size of the granular features is, in turn, determined from the starting powder size which must be sufficiently large to enable surface melting to be achieved, rather than near-instantaneous full melting of smaller particles. It has been suggested that the granular microstructure of the previously reported fibres is also responsible for the reported variability in fibre properties since any microstructural inhomogeneity becomes more evident when the microstructural features are of the order of the fibre diameter [28]. A similar granular microstructure in the fibres reported in the scale-up work here would also be a possible contributor, in addition to processing and control issues noted above, to the variability in fibre properties. If, on the other hand, the microstructure responsible for auxetic behaviour at 200ºC in the larger extruder is identified at the molecular chain level (nanoscale), then this has potential to lead to finer diameter and more uniform auxetic fibres than currently achievable from the microscale fused-particle microstructure at 159ºC in the smaller extruder. This would broaden the applicability of auxetic fibres to applications where the current fibres are too coarse (e.g. apparel and composite reinforcement).

4 Conclusion Auxetic fibres have been successfully produced on an industrial scale extruder for the first time. The fibres produced had good reproducibility and were found to exhibit auxetic response over a larger strain range than reported previously in laboratory-scale extrusions. There were some processing problems which will need to be further investigated in order to fully understand and commercialise the process. The processing parameters have been identified for this particular combination of powder and machine to be a barrel temperature of 200ºC, a screw speed of 12.5rpm and a take-up speed of either 1.5 or 3.5 rpm. Ambient air quenching of the extruded fibres was required.

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