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Extrusion Dwell Time and Its Effect on the Mechanical and Thermal Properties of Pitch/LLDPE Blend Fibres

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Abstract: Mesophase pitch-based carbon fibres have excellent resistance to plastic deformation (up to 840 GPa); however, they have very low strain to failure (0.3) and are considered brittle. Hence, the development of pitch fibre precursors able to be plastically deformed without fracture is important. We have previously, successfully developed pitch-based precursor fibres with high ductility (low brittleness) by blending pitch and linear low-density polyethylene. Here, we extend our research to study how the extrusion dwell time (0, 6, 8, and 10 min) affects the physical properties (microstructure) of blend fibres. Scanning electron microscopy of the microstructure showed that by increasing the extrusion dwell from 0 to 10 min the pitch and polyethylene components were more uniformly dispersed. The tensile strength, modulus of elasticity, and strain at failure for the extruded fibres for different dwell times were measured. Increased dwell time resulted in an increase in strain to failure but reduced the ultimate tensile strength. Thermogravimetric analysis was used to investigate if increased dwell time improved the thermal stability of the samples. This study presents a useful guide to help with the selection of mixes of linear low-density polyethylene/pitch blend, with an appropriate extrusion dwell time to help develop a new generation of potential precursors for pitch-based carbon fibres.

Keywords: blend; extrusion; dwell time; morphology; carbon-fibres; mesophase pitch; polyethylene

1. Introduction

Polymers are progressive substitutes for metal and wood, but their relatively poor performance regarding strength and stiffness, limits their ability to compete in many applications. Thus, in modern industries, from textiles to aerospace, there is an ongoing demand for improvements in the performance of polymer-based materials [1–3]. For example, there is great demand for carbon fibre (CF)-reinforced composite in the aerospace industry. Indeed, CFs using rayon as a precursor [4,5] were in widespread use in the early 1960s and, since 1963, pitch has successfully been used as the precursor for CFs with a superior elastic modulus [6].

The superior properties of CFs mean they are used in numerous applications from healthcare to space exploration [2]. Nevertheless, their industrial usefulness could be enormously improved if they were manufactured at lower cost and with enhanced mechanical properties [7]. Manufacturing CFs using petroleum derived pitch could reduce material costs for manufacturing CFs [8,9].

Previous reports [10] have shown that if the temperature is not controlled, the pitch filament could break in a brittle manner because the mesophase pitch fibre extrusion is sensitive to temperature changes. Researchers have also found that the fibres are also easily damaged during the spinning process and were difficult to handle before carbonization [11]. Other reports mention that decreasing the temperature by a few degrees significantly increased the viscosity of the mesophase pitch and increased the likelihood of brittle

fracture during winding [12]. Lim and Yeo (2017) reported that it is very hard to wind pitch fibres because they are brittle and have low tensile strength [13].

Aldosari et al. (2020) [14] showed how to obtain ductile pitch based carbon fibre precursors using pitch and LLDPE blend fibres, which were shown to be an appropriate material for manufacturing carbon fibres [15,16]. However, during the mixing of different materials/polymers the extrusion dwell time will have an effect on the physical properties of the final product [17,18]. The aim of this work is to systematically study the effects of extrusion dwell time on the mechanical and thermal properties of pitch and polyethylene blend fibres.

As a part of the current research, we varied the extrusion dwell time of LLDPE and pitch blend from 0 min to 10 min. The effect of the extrusion dwell time on the tensile strength, tensile modulus and onset degradation temperature was then determined experimentally. Such a study will be a valuable aid in the selection of appropriate dwell times for manufacturing ductile pitch-based precursor fibres for the future development of low-cost carbon fibre manufacturing.

2. Experimental Procedures

2.1. Materials

Bonding Chemical supplied the mesophase pitch, mesophase content 92% and Sabic supplied the LLDPE. Table 1 shows relevant properties.

Table 1. Relevant properties of MP precursor and LLDPE.

Relevant Property	MP Precursor	LLDPE
Softening Point	268 °C	99 °C
Melting point	298 °C	121 °C
Density	1.425 g/cm^3	0.918 g/cm^3

2.2. Materials Processing

Figure 1 shows a schematic flow chart illustrating the different factors affecting the screw extrusion process, which is used to blend consistent, uniform mixes, often of several materials [19]. To successfully investigate how to improve the material properties of the fibres it is necessary to appreciate how the mixing ratios and extrusion process affect the morphology of the fibres.
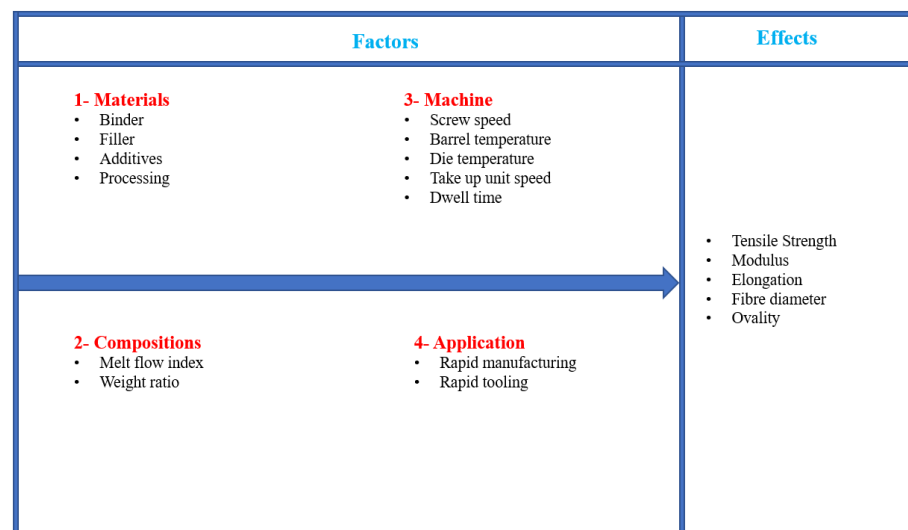

Figure 1. Effect of different factors on mixes of materials added to the extruder (adapted from Boparai et al. [19]).

A novel and valuable aspect of this work is that it completes the study of the interrelations between physical properties (both thermal and mechanical) and the microstructures of a linear low-density polyethylene/mesophase pitch (LLDPE/MP) blend over a range of extrusion dwell times.

"Mixing" is the processes whereby nonuniformity of concentrations are reduced. By definition, mixing increases the system's configurational entropy, which is a maximum when the locations of the constituents are random [20]. The time the polymer remains inside the extruder as a physically and chemically active hot melt will be termed the "effective dwell time". The distribution of pellets will determine their effective dwell time; the nearer the die the shorter the time, the nearer the hopper the longer the time Figure 2 [21].
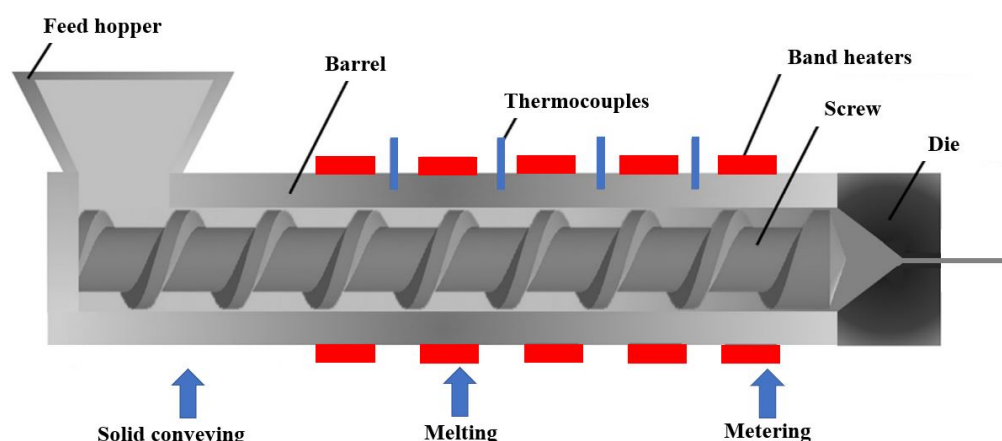

Figure 2. Extrusion of process of materials (adapted from Zhang et al. [22]). (Red blocks indicate band heaters).

The diameter of the nozzle of the Noztek extruder was 0.5 mm and its length was 1.5 mm, see Figure 2. The extrusion temperature is the temperature on the extreme right of Figure 2, at the entrance to the die was Figure 2. set to 315 °C with a volume rate of extrusion through the nozzle of close to 500 mm^3/min and a linear speed of 2.5 m/min, which was equal to the stretching speed. These values were maintained constant and the dwell time could be varied by changing the rate at which pitch and PE were fed into the extruder. The relative proportions to obtain the required fibre blend with 20 wt% LLDPE were, of course, constant. Figure 2 shows the stages within the extrusion process. The extrusion dwell times chosen to study the extrusion conditions for optimum thermal and mechanical properties of the CF were 0, 6, 8, and 10 min.

Note that a mix with 20 wt% LLDPE meant the mix contained 80 wt% mesophase pitch. All results presented below are for a blend of LLDPE/MP with 20 wt% of LLDPE unless otherwise stated.

3. Measurement Techniques

3.1. Scanning Electron and Optical Microscopy

The diameters of the fibres were measured using an optical microscope (a Nikon ECLIPSE ME600 at 20× magnification, Melville, NY, USA) with digital interference contrast microscopy to enhance sample contrast.

A Tescan VEGA3 SEM (Brno-Kohoutovice, Czech Republic) with Aztec software (Abingdon, UK) was used to investigate the specimens. They were prepared by being cut in 5.0 cm lengths, then having a thin layer of gold sputter-coated onto their end surfaces. For surface features, the specimens were placed lengthwise on horizontal aluminium stubs, and for examination of the end faces, were mounted end-on for analysis. For additional information, see [14].

3.2. Mechanical Tests

Tensile testing of the drawn and spun fibres were carried out according to ISO 11566-1996 [23], see Appendix A for Figure A1 and details of equipment used. A single filament was lightly stretched longitudinally across the centre of the elliptic slot, with the ends of the filament temporarily fixed using adhesive tape. With the filament in place, it was bonded to the mount using a single drop of Loctite 406 adhesive. To confirm repeatability and reproducibility, each fibre was the subject of six tests.

3.3. Differential Scanning Calorimetry and Thermogravimetric Analysis

The differential scanning calorimetry (DSC) measurements were made using a Mettler Toledo DSCQ2000 (New Castle, DE, USA) and thermogravimetric analysis (TA) using a Mettler Toledo Thermogravimetric Analyser TGAQ500 (New Castle, DE, USA). Both sets of measurements were performed in a nitrogen environment. The DSC samples were heated from 0 °C to 200 °C, then maintained at that temperature for 3 min and 20 s to remove all traces of any previous thermal events. The relevant thermal characteristics were found after carrying out cyclical heating and cooling of the specimens at a steady rate of 20 °C/min.

For the TA measurements, the samples were heated from 50 °C to 800 °C at 20 °C/min, then maintained at 800 °C for 5 min to remove any traces of prior thermal events.

4. Results and Discussion

4.1. Optical Microscopy

The morphology of the different blends of fibres was investigated using the Nikon system described above. As shown in Table 2, the diameter of the sample fibres increased with extrusion dwell time, see also Figure 3. Extrusion viscosity from the nozzle increases as temperature increases because the viscosity of the blend decreased. Our previous differential scanning calorimetry (DSC) data showed similar melting temperature for neat LLDPE and blend of pitch/LLDPE [14].

Table 2. Fibre diameter for LLDPE/MP as a function of extrusion dwell time.

Extrusion Dwell Time (min)	Fibre Diameter, μm
0	154 (±0.51)
6	189 (±0.26)
8	223 (±0.54)
10	234 (±0.30)

The amount of time that a randomly selected, small volume element in the feed stream spends inside the mixing equipment (extruder) is referred to as the residence or dwell time [24].

Shrinkage was observed in fibre diameter with an increase in LLDPE content for dwell time and all other factors kept constant. This was because fibre morphology changed as the LLPDE content increased, with the same extensive force producing a larger axial elongation [25]. Factors influencing the diameters of the extruded fibres included the viscosity of the polymer, die orifice diameter, and speed of extrusion.

4.2. SEM of LLDPE/MP Fibres

The images of the pitch fibres seen in Figure 4 show that microfibrils are present within fibres. In Figures 4a–h, and A2 the SEM images are for LLDPE/MP fibres with extrusion dwell times of 0 Figure 4a,b; 6 Figure 4c,d; 8 Figure 4e,f and 10 min Figure 4g,h.
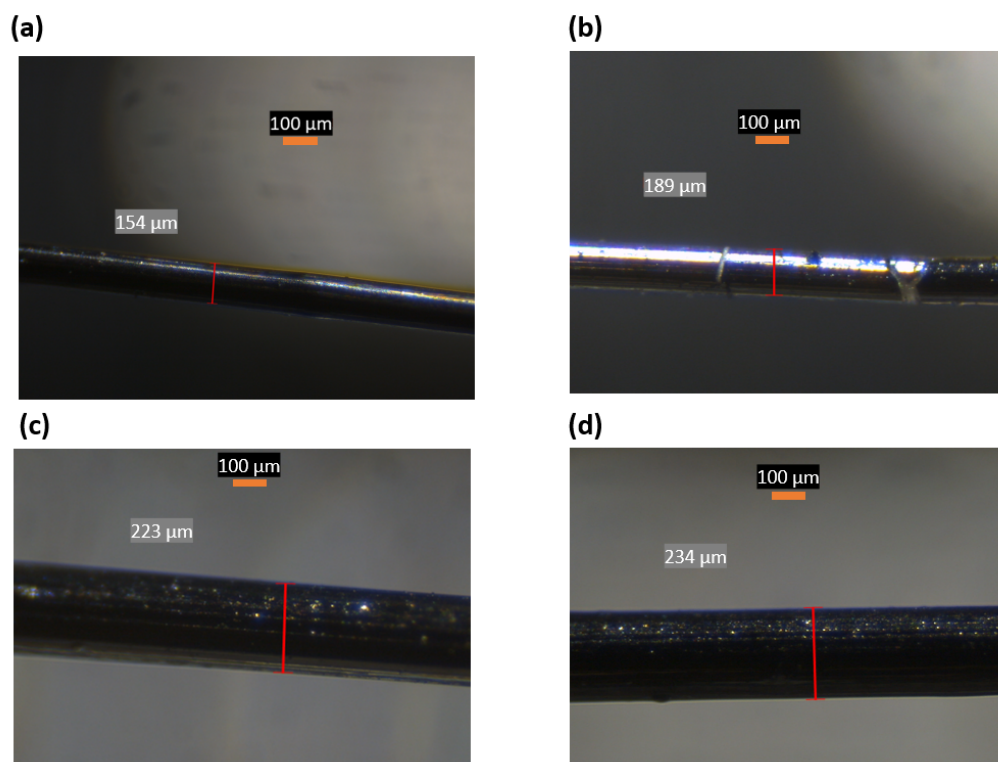
Figure 3. Images of LLDPE/MP fibres extrusion dwell times: (**a**) 0, (**b**) 6, (**c**) 8, and (**d**) 10 min.

There are two types of mixing: (1) distributive, or simple mixing, which refers to achieving a uniform spatial distribution of the different components, usually distributive mixing does not need high stresses, and (2) dispersive, or intensive mixing, which refers to achieving a fine level of dispersion and will often require reduction in the size of the components and so only occurs when the stress in the melt exceeds that necessary to rupture the component [21].

For zero minute dwell time the LLDPE pitch blend shows simple distributive mixing, which resulted in large mesophase pitch particles distributed in the LLDPE matrix Figure 4a,b.

However, with increases in dwell time from zero to 6, and then 8 min, distributive mixing of the pitch fibre in the LLDPE matrix was observed. By increasing the dwell time during the extrusion process from 0, to 6, to 8, and to 10 min the homogenous dispersion of pitch domain particles in the LLDPE matrix is increased Figure 4a–h. Thus, with a dwell time of 10 min, the matrix shows minimum number of voids; the pores trapped in the LLDPE/MP composite matrix had coalesced together. It may be, by increasing the extrusion dwell time, the diffusion bonding of LLDPE between the pitch domain is increased and reached its greatest value at 10 min. However, the 0, 6 and 8 min extrusion dwell times are not sufficient to form diffusion bonding between the fused LLDEP at the extrusion temperature of 315 °C.

The effect of the extrusion dwell time on the micrographs of the produced composite samples as shown in Figure 4 were determined by measuring the pitch domain diameters. Table 3 shows that the pitch domain size is reduced with increase in the extrusion dwell time. The MP molecules have a powerful inclination to align with the longitudinal axis of the fibre, a process which tends to start in the liquid crystalline phase [7].
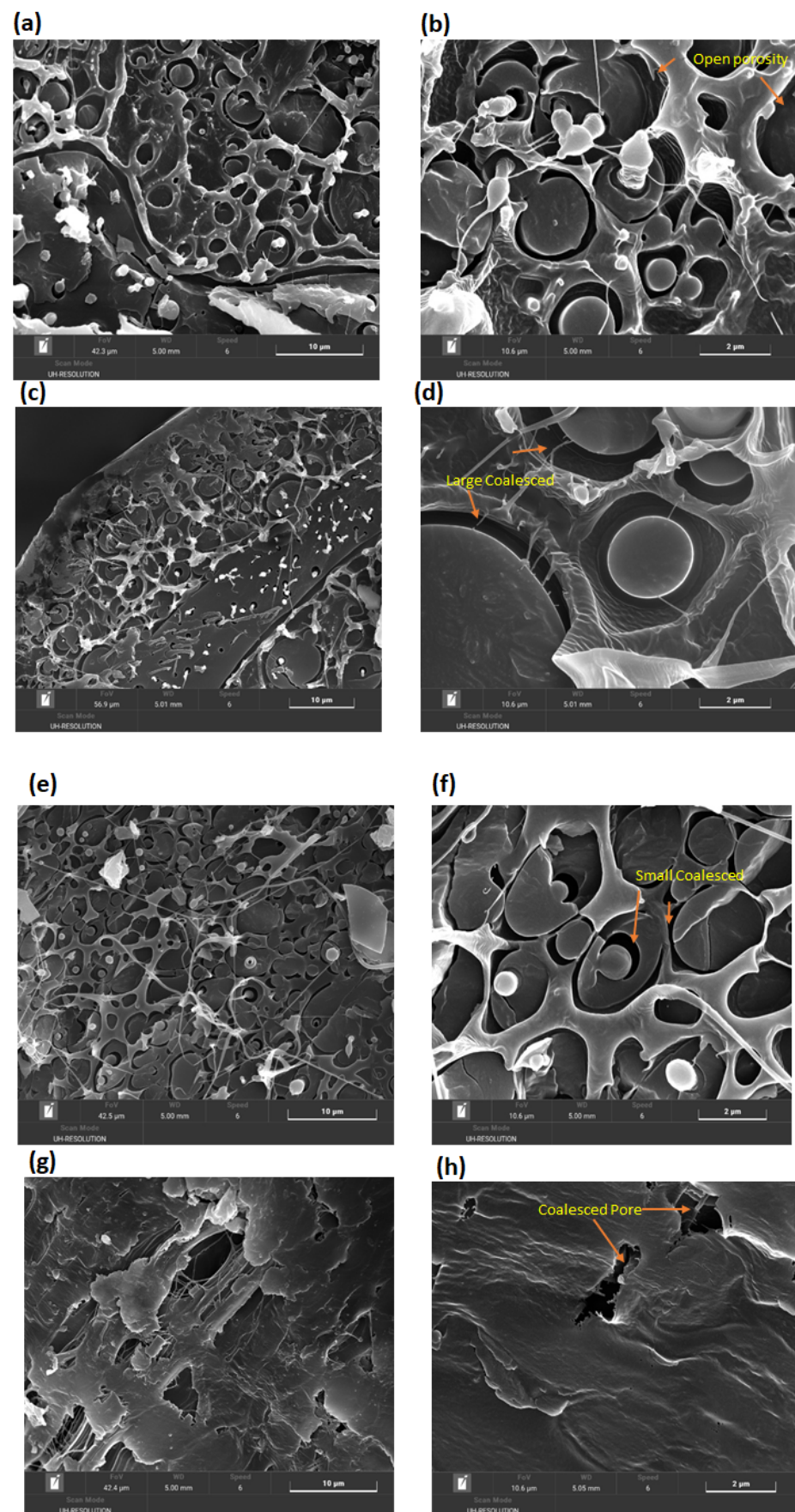
Figure 4. Morphology of LLDPE/MP fibres for different dwell times: zero (**a**,**b**), 6 (**c**,**d**), 8 (**e**,**f**), and 10 min (**g**,**h**).

Table 3. Particle size and air gap with extrusion dwell time.

Extrusion Dwell Time Duration (min)	Average Pitch Domain Size (μm)
0	1.38 (±0.2)
6	1.16 (±0.4)
8	1.08 (±0.3)
10	0.92 (±0.2)

4.3. Effect of Extrusion Dwell Time on the Tensile Strength

LLDPE is extensively used because it is inexpensive and versatile and has a tensile strength of about 6 MPa. This can be increased by adding fibre straws or multiple-walled carbon nanotubes to as high as 22 MPa [26–28], which would enable the uses of LLDPE to expand if it were to be used in combination with suitable additives.

The stress limit, the ultimate strength of extruded, neat LLDPE can be as high as 45 MPa, with strain of approximately 0.8, which implies that the LLDPE component within the fibres is critical for the load-bearing capacity of the LLDPE/MP fibres [14]. Figure 5 presents plots of stress against strain for LLDPE/MP fibres produced with different extrusion dwell times (Table 4). The samples showed high tensile strength (10.3 MPa) for zero dwell time but low strain to failure (0.23). However, samples with the highest extrusion dwell time (10 min) showed the lowest tensile strength (4.08 MPa) and highest strain to failure (0.60). We attribute this increase in strain to failure to the increased dwell time which allowed the LLDPE (the ductile polymer) to distribute more uniformly in the LLDPE/MP fibres and act as a plasticizer. Similar behaviour has been observed in epoxy rubber composites where the ductile rubber component helped to increase the strain to failure in the composite [29–33]. The samples with 6 min extrusion dwell time showed a good combination of high strength (8.36 MPa) and relatively high strain to failure (0.46). The results for an extrusion dwell time of 6 min appear anomalous because for this time we obtained moderate values of both ultimate strength and elongation, whereas for other durations we obtained either larger elongation values at the expense of ultimate strength, or higher ultimate strengths at the expense of elongation.
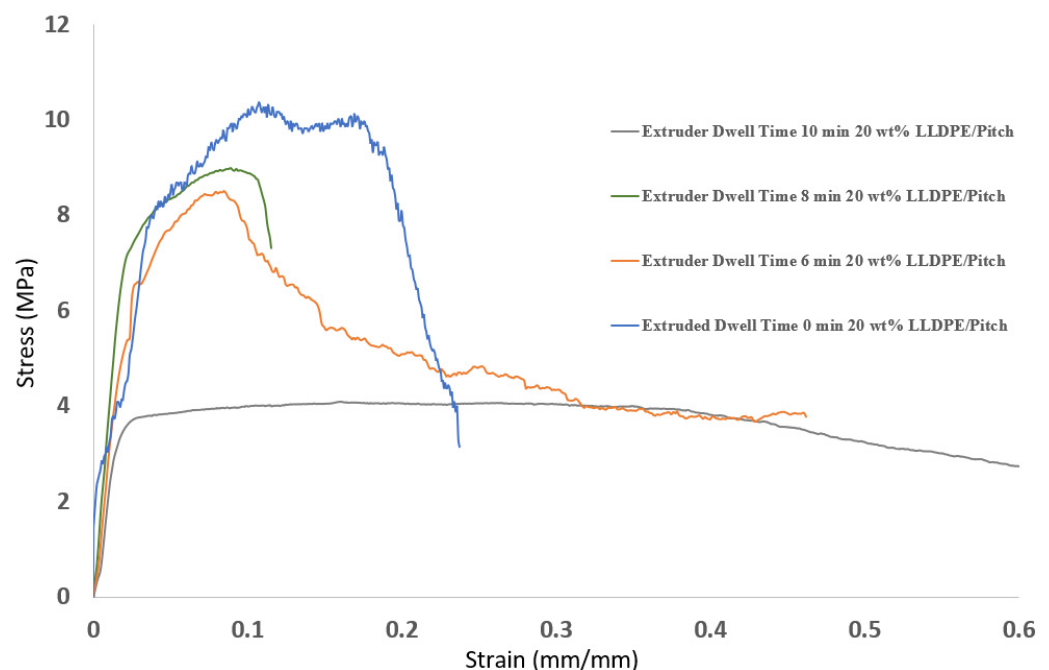

Figure 5. LLDPE/MP fibres, tensile strength of four different extrusion dwell times.

Table 4. Tensile strength, tensile modulus, and strain at failure of LLDPE/MP fibres for different extrusion dwell times. (Figures in brackets represent the standard deviation).

Extrusion Dwell Time (Min)	Tensile Strength (MPa)	Tensile Modulus (MPa)	Strain at Failure
0	10.3 ($\pm$0.87)	763 ($\pm$5.3)	0.23 ($\pm$0.025)
6	8.36 ($\pm$0.83)	823 ($\pm$4.5)	0.46 ($\pm$0.022)
8	8.98 ($\pm$0.57)	842 ($\pm$3.8)	0.11 ($\pm$0.036)
10	4.08 ($\pm$0.65)	857 ($\pm$5.6)	0.60 ($\pm$0.028)

4.4. Differential Scanning Calorimetry for LLDPE/MP Blend

As we have previously reported, Aldosari et al. [14] previously reported crystallization of different blends of LLDPE with LDPE and HDPE has been investigated using DSC. It was found that, typically, increasing the proportion of LLDPE, reduced crystallization temperature (usually ascribed to the LLDPE's higher molar mass), which increased chain entanglements, making crystallisation more difficult.

Analysis of the experimental results to obtain the required kinetic data revealed that the degradation rate of LLDPE was of relatively minor importance; the degradation started at 310 °C and 50% of the polymer degraded and was volatile at 430 °C [34]. The thermal properties of the given blend of LLDPE/MP with changes in extrusion dwell time between 0 and 10 min were investigated using DSC. Figure 6 shows the results of tests performed for a range of crystallization and melting temperatures.

For the given LLDPE/MP blend, Figure 6a,b presents well-ordered plots with clear crystallization peaks, showing that the more extended the extrusion dwell time the larger the peak value of the heat flow, except at 6 min. It is also observed, see Figure 6c,d, that for the given blend, increasing the extrusion dwell time increases the enthalpy of crystallization. Figure 6a,b also shows that the crystallisation temperature increased with increase in extrusion dwell time, confirming that the crystallisation temperature of the blend is affected by extrusion dwell time.

Figure 6c,d and Table 5 show that for the given LLDPE/MP blend, the melt temperature is a function of extrusion dwell time, as is the corresponding enthalpy of fusion. The melting or fusion temperature and maximum value of the enthalpy of fusion increased as extrusion dwell time increased. This is because the longer the extrusion dwell time the fewer crystalline spheres are present in the blend [14], confirming that the extrusion dwell time influenced both the fusion and crystallisation temperatures of the blends. The effect of extrusion dwell time on the fusion and crystallization temperature is negligible at all extrusion dwell times and the percentage difference in their values between maximum and minimum time is less than 1%. On the other hand, the extrusion dwell time has a more pronounced effect on the enthalpy of fusion compared with fusion and crystallization temperature. Therefore, the percentage difference in the enthalpy of fusion between 6 and 8 min is equal 37.5%. On other hand, the percentage difference between 6 and 10 min is equal 50% as shown in Table 5. This means that the effect of extrusion dwell temperature is more pronounced on the enthalpy of fusion compared to fusion and crystallization temperature by increasing the dwell time.

Table 5. Fusion and crystallisation temperatures of LLDPE/MP blend.

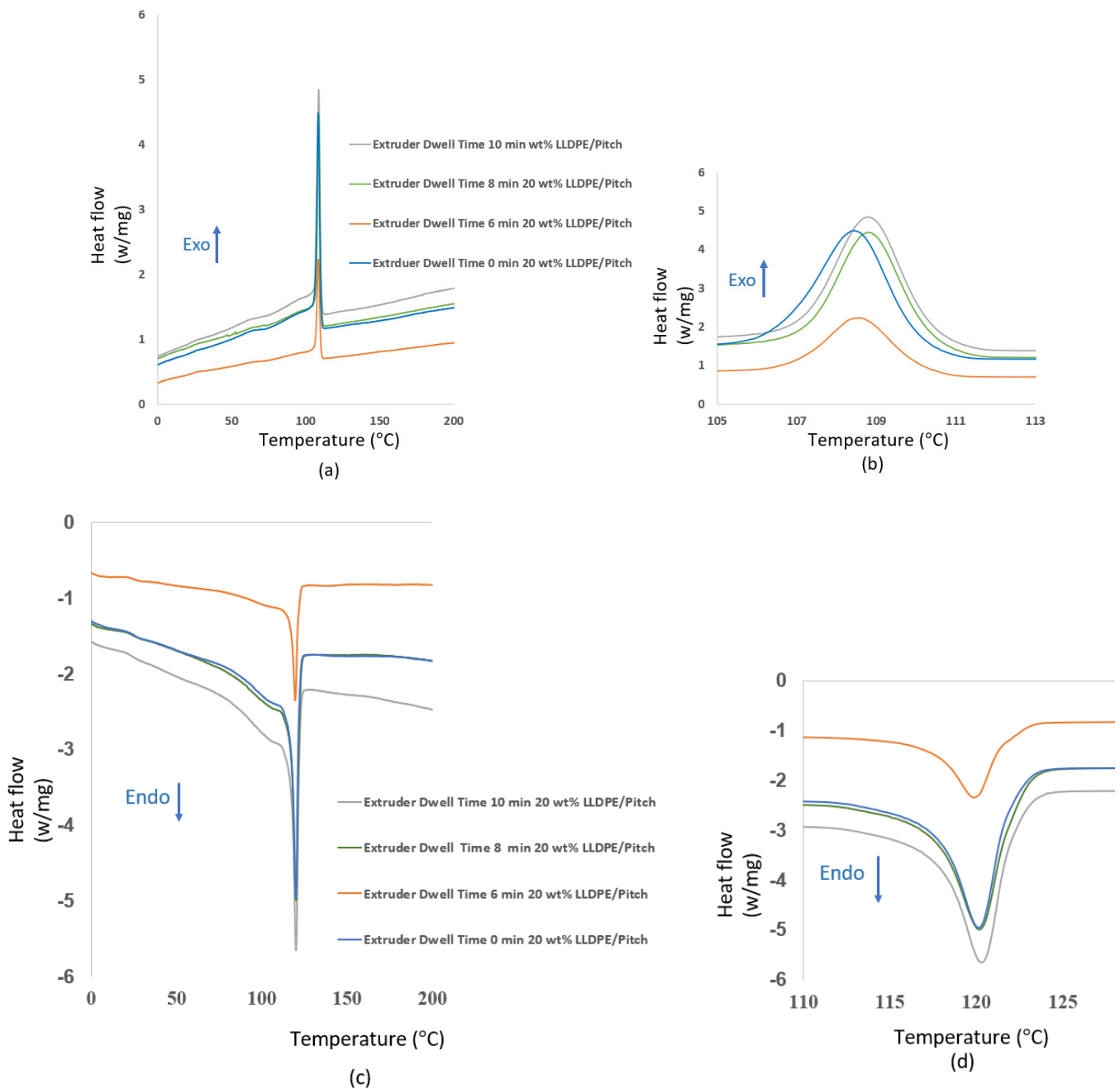| Extrusion Dwell Time (min)	Fusion Temperature (°C)	Crystallization Temperature (°C)	Enthalpy of Fusion (kJ/mol)	Enthalpy of Crystallisation (kJ/mol)
0	123.0	102.0	350	1750
6	122.8	101.6	271	1355
8	123.5	102.7	372	1860
10	123.6	102.9	406	2030

Figure 6. (**a,b**) Exothermal heat flow vs. temperature for crystallisation of LLDPE/MP sample and (**c,d**) endothermic heat flow for melting of LLDPE/MP sample. Four different extrusion dwell times. (Enthalpy of crystallisation, ΔH_c, and enthalpy of fusion, ΔHm, are the areas under the respective curves).

DSC was used to assess the crystallization temperatures of the LDPE and LLDPE blend for temperatures between 110 °C and 120 °C, a temperature range in which pure LDPE does not melt. The population of crystallites melting increased with a 10 °C increase in temperature [35]. The longest extrusion dwell time (10 min) had the highest level of crystallization, possibly due to the influence of dwell time on crystallization during the extrusion process.

The enthalpy of fusion and melt temperature of the given LLDPE/MP blend changed with extrusion dwell time. It was observed that the longest dwell time (10 min) had the highest value for the melt temperature at 123 °C which may be due to the high crystallinity of the sample subject to the 10-min dwell time.

4.5. Thermogravimetric Analysis of the LLDPE/MP Blend

It is reported in the literature that higher decomposition temperatures of composite materials mean greater thermal stability [28,36,37]. Thermal decomposition for the given LLDPE/MP blend was investigated for different extrusion dwell times, and the resulting TGA curves, loss of mass vs. temperature, can be seen in Figure 7. The analysis was performed for temperatures from 100 °C to 800 °C.
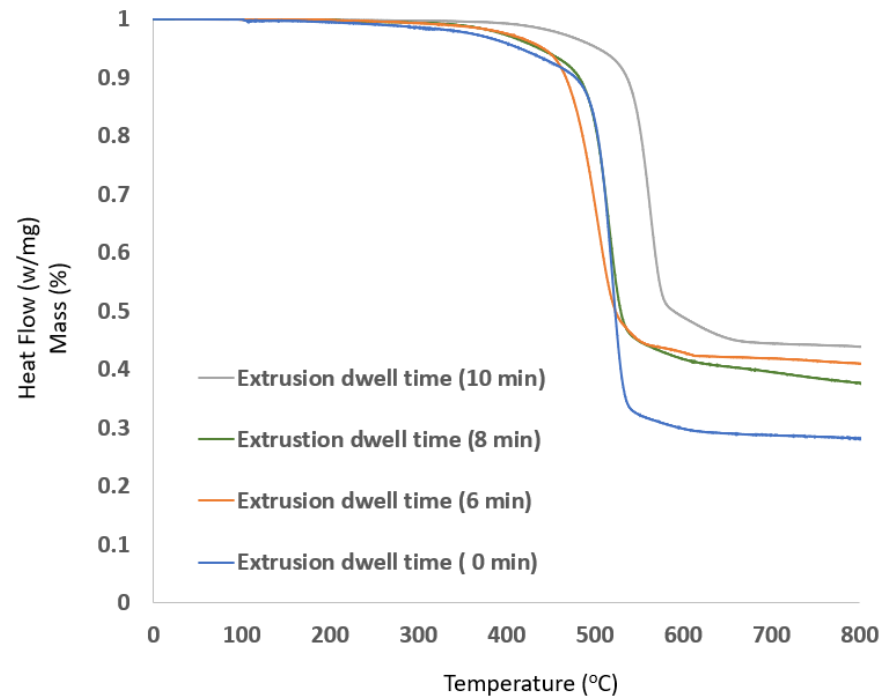

Figure 7. TGA curves for different extrusion dwell times for given LLDPE/MP blend.

Decomposition temperatures for various extrusion dwell times are shown in Figure 7 and Table 6. Those fibres subject to 10 min extrusion dwell time preserved more than 55% of the fibre mass until about 800 °C. For zero extrusion dwell time only about 30% of the mass was preserved, while for 8 min extrusion dwell time about 40% of mass was preserved. This confirms that, generally, the longer the extrusion dwell time the more thermally stable the blend, i.e., that the duration of the extrusion dwell time affects thermal stability. However, we also see that the mass preserved for 6 min extrusion dwell time can surpass that preserved for 8 min. With their decreased brittleness and relatively higher thermal stability, the 8 and 10 min extrusion dwell times showed higher onset degradation temperatures compared the other sample times and could provide acceptable precursors for the production of CFs. The onset points of decomposition that occur at which the sample shows a loss of 1 wt.% of its initial mass as shown in Table 6 due to the onset degradation temperature increased by increasing the extrusion dwell time.

Table 6. Onset and final degradation temperatures for LLDPE/MP with extrusion dwell time.

Extrusion Dwell Time (min)	Onset Degradation Temperature * (°C)	Final Degradation Temperature (°C)	Final Residue (%)
0	492.7	576.4	31
6	485.3	609.5	43
8	501.1	598.4	40
10	553.2	634.4	45

* Onset degradation temperature: temperature at which the sample has lost 1% of its initial mass.

5. Conclusions

This paper has shown that the extrusion dwell times for a LLDPE/MP blend (with 20 wt% LLDPE) can enhance the fibre morphology, mechanical and physical properties. For a range of extrusion dwell times from zero to 10 min, it has been shown that increasing the time significantly affects the morphology of the LLDPE/MP blend. It has also been demonstrated that the diameter of the fibres and the enthalpy of fusion both increased by increasing the extrusion dwell time due to die swell.

TGA results demonstrated that, generally, by increasing the extrusion dwell time the onset degradation temperature is increased and the final degradation temperature increased the residual mass (the mass remaining constant after 630 °C). Hence, optimisation of extrusion dwell time should be taken into account when considering the thermal stability and mechanical performance of LLDPE/MP blend fibres. An extension dwell time to 6 min showed a useful combination of high tensile strength, high strain to failure and relatively good thermal stability. Mechanical tests on the given LLDPE/MP blend showed that, generally, increasing the extrusion dwell time increased strain and enhancing failure.

It is expected that the findings reported in this work will be valuable to the development of a new generation of pitch-based carbon fibre precursors for end application in the aerospace industry.

Author Contributions: S.M.A. and S.R. conceptualized the theory and the method. S.M.A. performed the experimentation and the analysis. All authors have read and agreed to the published version of the manuscript.

Data Availability Statement: Data can be available on a request from the corresponding author via email.

Appendix A

The tensile test procedure was conducted according to the standard ISO 11566-1996.
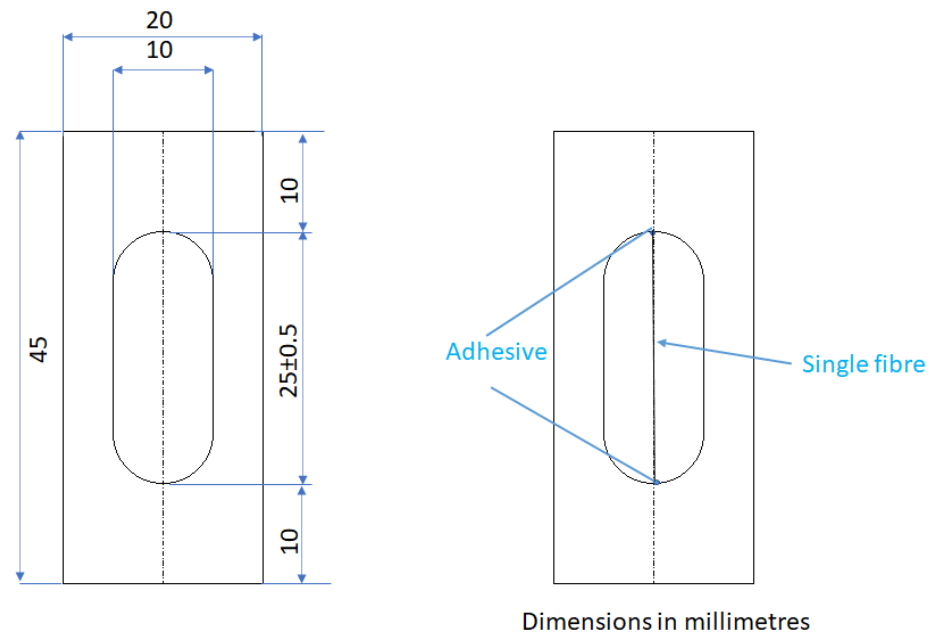

Figure A1. Tensile test setup for melt spun extrusion dwell time MP/PE fibre using a DEBEN Microtest fibre tensile tester and a Leica EC4 microscope.
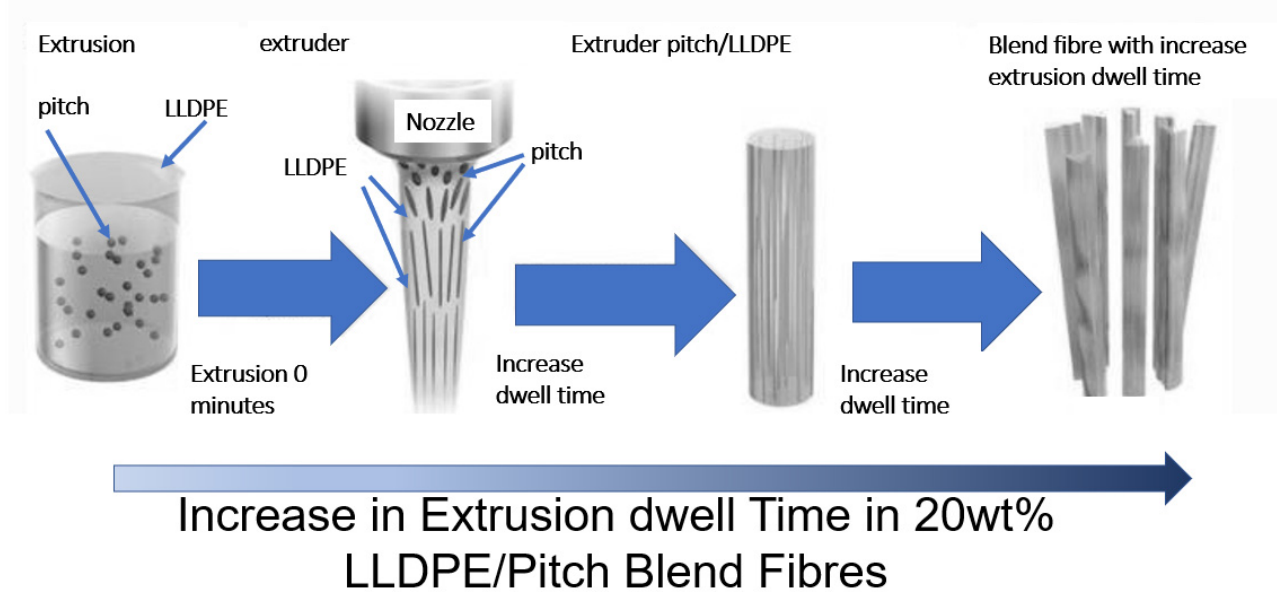
Figure A2. Increase in extrusion dwell time in 20 wt% LLDPE/pitch blend fibres.

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