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Tensile Properties of Extruded Short Glass Fibre/Low Density Polyethylene Composites

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A B S T R A C T

Home-made single screw extruder was designed and constructed with screw length to diameter ratio 20:1 operating at different screw speeds ranging from 7-28 rpm and at different temperatures. The extruder was used to produce rod composite samples composed of low density polyethylene and short E-glass fibre. The effects of fibre contents and extruder operating variables on the tensile modulus and ductility were investigated. The tensile modulus increased with fibre weight percent up to 10.52 wt% as residual fibre content (20 wt% prepared sample before processing) and had a value of 827 MPa, then slightly decreased to lower values due to bad wettability and poor distribution/dispersion of fibres in the LDPE matrix. The clear decrease in fibre content after processing (residual fibre content) which is about 40-55% is due to the sticking of fibres in hopper or in the different extruder zones. On the other hand, ductility as percentage elongation-at-break decreased significantly at low fibre concentration then decreased in lower rate at higher fibre contents. Operating variables of the extruder were found to affect the tensile properties of LDPE/glass fibre composites by affecting the interfacial adhesion between the two components. This effect was clear from the analyzed data of the modulus efficiency factor K_E.

Key Words:

glass reinforced composites;
tensile modulus;
modulus efficiency factor;
ductility;
residual fibre content.

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INTRODUCTION

Low density polyethylene is a non-toxic thermoplastic polymer used in polymer industry and is one of the most versatile polymers due to its low density, low cost, and high processability. However, its use is restricted because of several drawbacks, such as low strength, low tensile modulus, and poor heat resistance [1-4]. Mechanical properties of polyethylene can be modified and enhanced by adding different modified additives such as glass beads, calcium carbonate, talc, mica, wood fibres, sisal, kenaf, pineapple, etc. Most of the incorporation of short glass fibres which increases the stiffness, strength, and thermal stability of polyethylene [3,4].

Short fibre reinforced thermoplastics are very important commercial materials. Where a good processability is typical of short fibre composites, the mechanical properties of final products mainly depend on several factors such as fibre content, inherent properties of fibres and polymer matrices, fibre surface treatment and impregnation, adhesion and interaction of fibres into the polymeric matrix, residual fibre length, and orientation and distribution of fibres within the polymeric matrix. Some of these factors are affected by the processing technique used to manufacture the reinforced thermoplastic polymers [5-7].

Extrusion has become the most popular process for compounding the dispersed or short fibre reinforcement with a thermoplastic mainly due to its economic and continuous operation. Single screw extruders with a screw length to diameter ratio of 20:1 or above find application in blending chopped glass fibres with thermoplastic polymers [8].

Tensile properties of short fibre reinforced composites depend on the orientation of fibres within the matrix (i.e., longitudinal or random) and can be estimated theoretically by equations which are derived from Halpin–Tsai equations [9]. The equations which are applied to predict the modulus of the composite are as the follows:
longitudinal modulus:

$$E_L = \frac{1+2(L_f/d_f)\eta_L V_f}{1-\eta_L V_f} V_m \quad (1)$$

transverse modulus:

$$E_T = \frac{1+2\eta_T V_f}{1-\eta_T V_f} \quad (2)$$

where,

$$\eta_L = \frac{(E_f/E_m)-1}{(E_f/E_m)+2(L_f/d_f)} \quad (3)$$

and

$$\eta_T = \frac{(E_f/E_m)-1}{(E_f/E_m)+2} \quad (4)$$

where, L_f, d_f, V_m, V_f, E_m, E_f are fibre length, fibre diameter, volume percent of matrix, volume percent of fibre, tensile modulus of matrix, and tensile modulus of fibre, respectively.

Also Tsai developed a method of calculating the tensile modulus of composite which are randomly oriented in 2-D.

$$E_{2D} = \frac{3}{8}E_L + \frac{5}{8}E_T \quad (5)$$

Fibres may also be oriented randomly in three dimensions to give truly isotropic materials. An approximate equation for modulus in this case is given by eqn (6) [10]

$$E_{3D} = \frac{1}{5}E_L + \frac{4}{5}E_T \quad (6)$$

where, E_L and E_T are estimated from eqns (1) - (4). Tensile modulus depends on the interaction between fibres and matrix and it is also affected by fibre aspect ratio and orientation which can be related to modulus efficiency factor K_E estimated from the modified rule of mixture [11-12]

$$E_C = K_E V_f E_f + V_m E_m \quad (7)$$

modulus efficiency factor can be related to Cox's fibre length efficiency factor η_l and orientation efficiency factor η_0 [13]. Values of K_E and the degree of discrepancy between the theoretical and experimental tensile moduli indicate the compatibility and adhesion between fibres and polymeric matrix.

Another property which is important to be estimated is ductility. Ductility can be defined as the ability of material to withstand plastic deformation up to fracture and is estimated in terms of percentage elongation-at-break as follows [14]:

$$Ductility\ (El\ \%) = \frac{l_f - l_o}{l_o} \times 100\% \quad (8)$$

where, l_o and l_f are initial length and final length of sample at break, respectively.

In this work, home-made single screw extruder is used to process a composite material composed from low density polyethylene as a polymeric matrix and chopped strand short E-glass fibre as reinforcement.

The effects of fibre content on modulus and ductility have been observed. The extruder was operated at different temperatures and screw speeds to determine their effects on the orientation of fibres, wettability, and aspect ratio of the reinforcement which have a direct effect on the tensile modulus and ductility of composite materials. Modulus efficiency factor and theoretical values of tensile moduli were estimated to indicate the compatibility and adhesion between fibres and LDPE matrix. Thus, the novelty of this work is the concept of modulus efficiency factor and its dependency on the above parameters and factors that affect the mechanical properties of the composites. On the other hand, examining the effect of mixing technique used prior to the extruder on the fibre content is one of the objectives of this work.

EXPERIMENTAL

Materials

The material used in this research was low density polyethylene. IPethen ®323 LDPE supplied by Carmel olefins Company with melt flow reference (190; 2.16) of 2g/10min. Short E-glass fibres in the form of chopped strands of about 38 mm in length and 10.6 μm in diameter for each fibre supplied by VOSS-CHEMIE were used at different concentrations. The fibre length was reduced to 8 mm before processing.

Processing

Low density polyethylene and short glass fibres were mixed together at different weight percentages (5, 10, 15, 20, 25, 30, 35, 40 and 45) by using a simple tumbling device. The blends were introduced into the feed zone of the extruder. The extruder had a L/D ratio of 20 with three distinct zones (feed, compression, and metering). The flight depth in the metering zone was 2.25 mm, helix angle 17.7º, which could be operated at different rotating speeds varied from about 7 to 28 rpm. The extruder was attached with a capillary die of 10 mm in diameter. The prepared samples were extruded at the same temperature 200ºC and the same screw speed of 17 rpm. The samples which emerged from the die were cooled to room temperature by a water path. A 20 wt% sample has been processed at different temperatures (180, 200, and 220ºC) and at different screw rotation speeds (7, 17, and 28 rpm) to examine the effect of processing variables on the mechanical properties of the composite.

Testing

Residual Fibre Content

This test was carried out examine the effect of processing technique used on the residual fibre content in the polymeric matrix. Five samples from each prepared composites (5, 10, 15, 20, …, 45 wt%) were placed in a furnace at 450ºC for 5 h [5] to digest the LDPE matrix and leave fibres alone. The remaining fibre was weighed and the residual fibre weight percentage was calculated.

Tensile Test

Tensile test was carried out by using Gunt Hamburg apparatus WP 310 machine at constant speed 5mm/min and at room temperature. Five trials of each sample were taken. The samples were 100 mm gauge length and 6 mm gauge diameter. Tensile test was done to measure the tensile modulus and ductility (as percentage elongation).

Morphological Test

This test was performed by using an optical microscope BX 60 M system Microscope Olympus. It was done to observe the break down of the fibres and measure the residual fibre length after processing, for that the matrix should be digested at 450°C for about 5 h to burn the low density polyethylene matrix and just leave the reinforcement component. The residual fibre length is measured under the microscope by using an attached micrometer, the length is reported as the average length of ten segments taken randomly from the digested samples.

RESULTS AND DISCUSSION

Residual Fibre Content

Fibre content found to be decreased after processing with an average weight percentages (2.33, 4.57, 7.35, 10.52, 14.44, 17.82, 23.25, 28.35, and 33.18 wt%). This decrease is due to the sticking of fibres in the hopper or remaining in the extruder zones, because of the inefficient method of mixing used prior to the extruder. To improve the efficiency of the designed extruder and/or prevent the sticking of fibres and the decrease in the residual fibre content; compounding machines (suitable mixers) may be used prior to the main operation. Compounding the fibres into the polymeric matrix would lead to well distributive/dispersive mixture and the properties of the final composite are improved.

The Effect of Fibre Content on Tensile Modulus and Ductility

Elastic modulus was calculated from the tangent at low stresses for the elastic region in the stress/strain curve. To examine the degree of compatibility between the fibres and the matrix, theoretical values of the composite tensile modulus were calculated in three different manners according to the orientation of fibres within the LDPE matrix; in one direction (longitudinal short fibre), randomly oriented in two dimensions, and randomly oriented in three dimensions. Theoretical tensile moduli were calculated, which were based on the residual fibre content and residual fibre length equal to 4 mm.

Figures 1, 2 show the relation between fibre content as weight percentage of the experimental and theoretical tensile modulus, respectively. It is clear from Figure 1 that the tensile modulus increases with fibre content up to 10.52% residual fibre content (20 wt% prepared sample) then decreases slightly to a lower

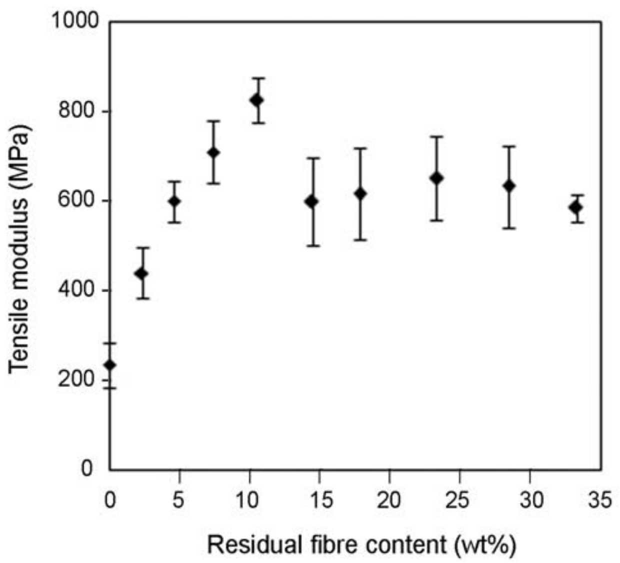

Figure 1. Relation between experimental tensile modulus and residual glass fibre content.

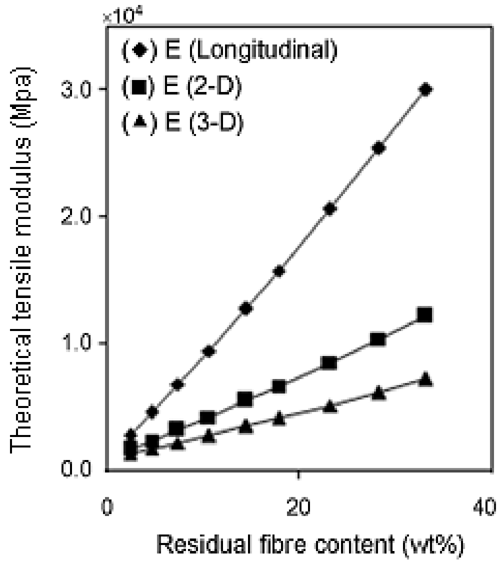

Figure 2. Relation between theoretical tensile modulus and residual glass fibre content.

value and remains nearly constant. It is believed that this decrease is due to the decrease in wettability of the matrix to the fibres, where the matrix loses its ability to wet the fibres at high concentration and therefore, voids are formed in the composite and fibres act alone and the matrix cannot transfer the load between the fibres that are not impregnated.

Another reason for this unexpected decrease after such low fibre content may be due to poor distribution and/or dispersion of glass fibres within the low density polyethylene matrix which is related to the use of inefficient method of mixing. The increase of tensile modulus with different reinforcements for thermoplastic composites has been observed by several researchers [3, 15-17]. The values of tensile modulus are very low compared with the theoretical ones, this discrepancy may be due to the damage of fibres during processing, poor distribution /dispersion of fibres in the polymeric matrix, or it can be related to the poor adhesion between glass fibre and low density polyethylene. As we believe, the interfacial adhesion between fibres and matrix can be improved by the use of appropriate coupling agents which must be compatible with both the reinforcement and the polymeric matrix such as a suitable silane or copolymer. Where, poly (ethylene-*g*-maleic anhydride) is a suitable copolymers used for this purpose as reported by other researchers [4]. Poor adhesion can be observed

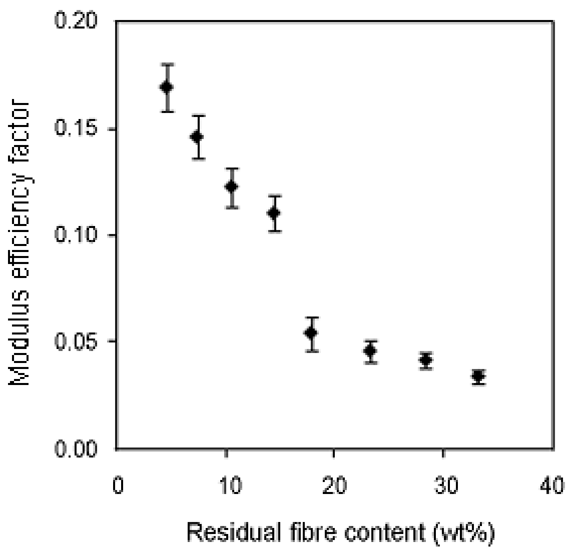

Figure 3. Effect of residual fibre content on modulus efficiency factor.

by the estimation of the modulus efficiency factor K_E from the modified rule of mixture. K_E has very low values and decreases with increasing fibre contents as it is illustrated in Figure 3.

Percentage of elongation-at-break value was estimated to observe the effect of fibre content on the ductility of the polymeric matrix. This effect is shown in Figure 4. It is obvious that Figure 4 exhibits a significant decrease in ductility at lower fibre concentration where, its value decreases at lower rate. This behaviour may be attributed to the formation of dense structures within the composite volume as the fibre concentration increases, leading to the production of anisotropic final product. Gergopoulos et al. [3] and Gupta et. al. [16] have found the same behaviour for other thermoplastic fibre composites. Also, Bkiaris et al. [4] have found similar trend for glass fibre/LDPE composite before adding silanes or copolymers.

The Effect of Extruding Temperature on the Tensile Modulus and Glass Reinforced LDPE Processed at Constant Screw Speed (17 rpm)

The effect of temperature on tensile modulus is illustrated in Figure 5. The 20 wt% composite samples were prepared at different temperatures (180, 200, and 220°C) to observe their impact on the properties of the final product. Viscosity of most polymer melts decreases with increasing temperature [18]. This decrease in viscosity affects the wettability of the matrix to the fibres and it may also affect the dispersion of reinforcement within the polymeric matrix. All these aspects have direct effect on the mechanical properties of the reinforced polymers.

As it is shown in Figures 5, tensile modulus at 180°C has lower values than those at 200°C. This can be attributed to a decrease in the viscosity of LDPE matrix and in this case liquid (matrix) will flow over the reinforcement, covering greater areas of the rough surface while displacing more air. This means that at

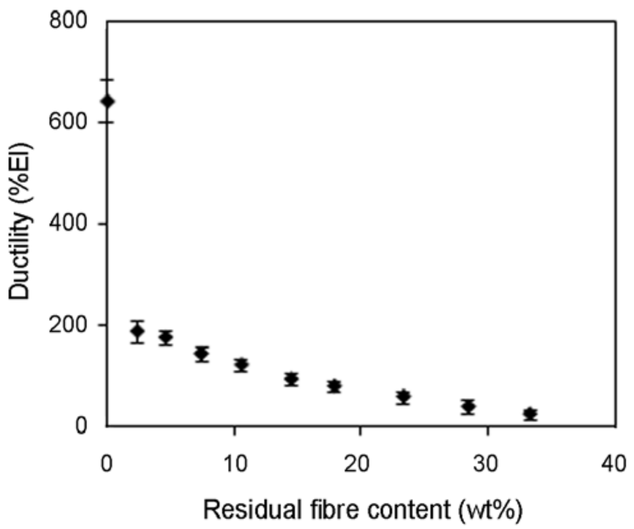

Figure 4. Effect of residual glass fibre content on ductility as percent elongation-at-break.

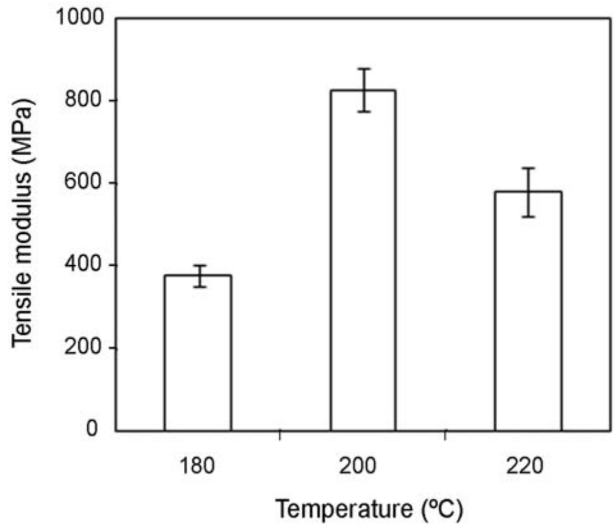

Figure 5. Effect of processing temperature on tensile modulus of glass fibre/LDPE composite.

Table 1. The effect of processing temperature on adhesion between glass fibre and low density polyethylene.

Temperature (°C)	Wettability	Orientation	K_E	Adhesion
180	Bad	Good	Very low	Very poor
200	Fair	Good	High	Good
220	Good	Bad	Low	Poor

200°C, there are considerable wettability and adhesion compared to 180°C. Also, it is valuable to notice that there is no change in the orientation and distribution of the fibres within the polymeric matrix.

On the other hand, at 220°C tensile modulus decreases although there is an increase in wettability of the fibres and the matrix. This decrease is due to the loss of fibres longitudinal orientation and distribution within the low viscosity polymeric matrix which is resulted from the large plastic flow that occurs in the matrix (presented by Sanomura et al. [13]) or to some extent to the degradation of the matrix at high temperatures.

Wettability and orientation are the factors which affect the degree of adhesion between glass fibre and LDPE which can be estimated by the modulus efficiency factor K_E. For the composite samples prepared at different temperatures K_E is shown in Figure 6. The effect of processing temperature on the adhesion between reinforcement and low density polyethylene is summarized in Table 1.

The Effect of Screw Speed on the Tensile Modulus of Composite Processed at Constant Temperature (200ºC)

Speed of rotation has a significant effect on both the polymeric matrix and the reinforcement. Shear rate applied on the processed material increases by increasing the speed of rotation. The matrix viscosity decreases because polymer melt acts as a shear thinning (pseudoplastic) fluid [18]. Increasing the shear rate will damage the fibres, decrease their residual length, and change orientation from longitudinal to 2-D or 3-D randomly oriented composite. All these parameters, viscosity, residual fibre length, and orientation affect the tensile modulus and ductility of the short glass reinforced LDPE. The effect of screw speed on tensile modulus of 20wt% prepared samples

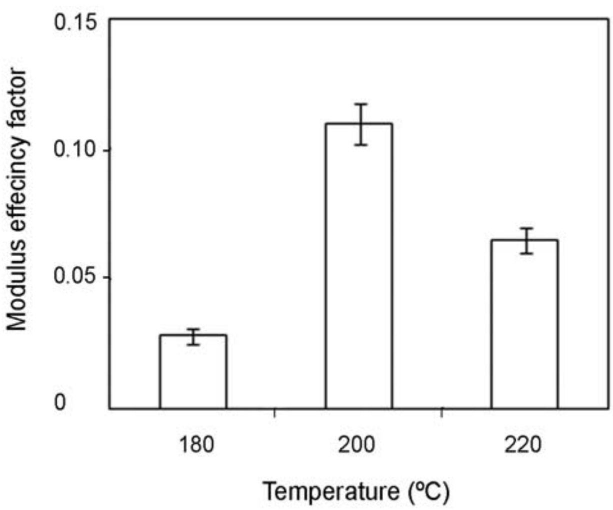

Figure 6. Effect of processing temperature on modulus efficiency factor.

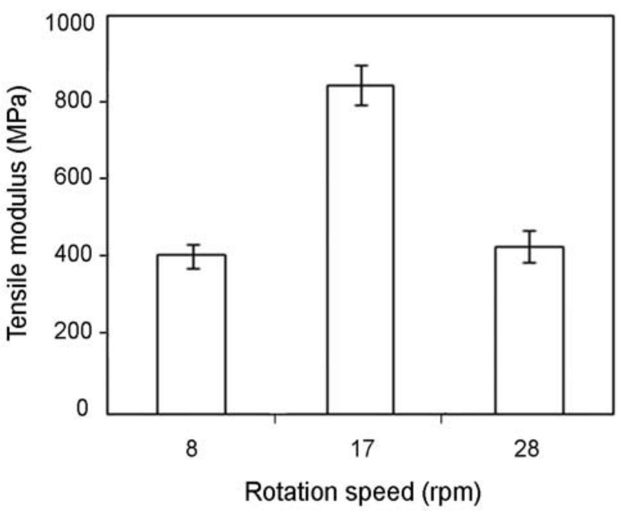

Figure 7. Effect of screw speed on tensile modulus of glass fibre/LDPE composite.

Table 2. The effect of screw speed on adhesion between glass fibre and low density polyethylene.

Speed (rpm)	Fibre length (mm)	Wettability	Orientation	K_E	Adhesion
8	5.2	Bad	Good	Very low	Very poor
17	4	Fair	Good	High	Good
28	2.8	Good	Very bad	Low	Very poor

is shown in Figure 7.

Residual fibre length depends on the parameters of the processing techniques. In this work, it was found that the residual fibre length decreased with speed of rotation. Before processing, the average fibre length was about 8 mm which was decreased to 5.2, 4, and 2.8 mm at 8, 17, and 28 rpm, respectively. Effect of parameters of processing techniques as screw speed or extrusion rate were studied by other researchers and they have found that screw speed has nearly the same effect on the fibre residual length [5,13,19-20].

It is obvious from Figure 7 that the speed of rotation affects tensile modulus of reinforced LDPE. High values were obtained at moderate speed of 17 rpm. At low speed there is low shear rate which reduces the average fibre length from 8 mm before processing to 5.2 mm. This condition keeps fibres in good orientation and low shear rate would give high melt viscosity. Therefore, the matrix cannot wet the fibres which in spite of good fibres orientation it leads to bad and poor adhesion of the LDPE and glass fibre. This poor adhesion produces low values of tensile modulus.

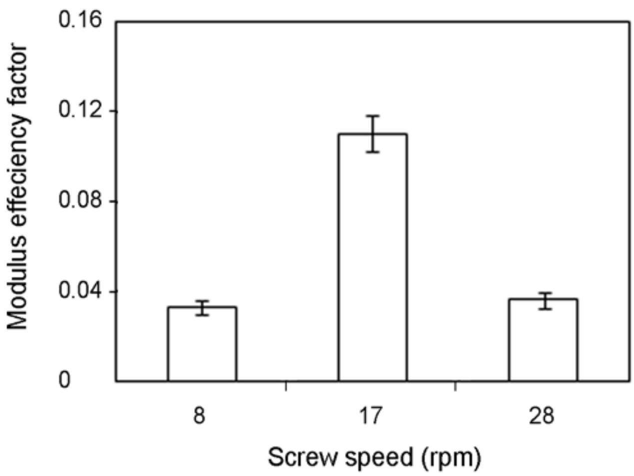

Figure 8. Effect of screw speed on modulus efficiency factor.

On the other hand, at high speed of 28 rpm, it is noticed that tensile modulus and ductility decrease again. This phenomenon can be attributed to the great damages of the fibres where fibre length reduced from 8 mm before processing to about 2.8 mm and also fibres being oriented randomly. These results can be supported by the estimation of modulus efficiency factor, which depends directly on wettability, orientation, and aspect ratio. This is an indication of the degree of adhesion between LDPE matrix and short glass fibres. The effect of screw speed on modulus efficiency factor is shown in Figure 8 and its effect on adhesion is summarized in Table 2.

CONCLUSION

Home-made single screw extruder with designed parameters can be used to process short glass reinforced thermoplastics. Optimum parameters of operating conditions; temperature, and screw rotation speed led to high wettability, longitudinal orientation of the fibres within the matrix, and minimum damage of the fibres. All these parameters were related to modulus efficiency factor K_E which is a direct indication of the adhesion between fibres and the matrix. It was concluded that if the extruder operated at 200°C and its screw rotated at moderate speed, better adhesion would be obtained and the best mechanical properties of the final product are achieved. It is noticed from the previous results that tensile modulus of LDPE-short glass fibre composite processed through the designed extruder increases up to 10.52 wt% residual fibre content (20 wt% prepared sample). This is then decreased to relatively lower value due to the formation of voids and the decrease in wettability between the fibres and matrix which lead to the decreased adhesion. It is clear that there is a great discrepancy between the values of experimental and theoretical tensile moduli which may be explained by the damage of fibres, poor distribution/dispersion of fibres in LDPE matrix, and poor adhesion between reinforcement and matrix. From the values of the residual fibre weight percentages (fibre content after processing) it is concluded that the mixing method used prior to the extruder is inefficient and leads to the decrease in fibre content and the poor distribution /dispersion of fibres within the polymeric matrix.

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