INVESTIGATION OF INTERFACIAL STRENGTH BETWEEN POLYETHYLENE TEREPHTHALATE FIBRE AND MALEIC ANHYRDIDE MODIFIED POLYPROPYLENE

F. L. Gentles[1*], J. L. Thomason[1]

[1] *Department of Mechanical and Aerospace Engineering , University of Strathclyde, 75 Montrose Street, Glasgow, Scotland, UK, G1 1XJ*
** Fiona.gentles@strath.ac.uk*

Keywords: Polyethylene Terephthalate Fibre, Polypropylene, Interfacial Strength, Single Fibre Pull Out

Abstract

Single fibre pull out was used to investigate the interfacial shear strength (IFSS) between Polyethylene Terephthalate (PET) fibre and Polypropylene (PP) matrix containing various percentages of maleic anhydride (MA). Glass Fibre (GF) – PP was used as a benchmark for the PET-PP IFSS to ascertain whether PET fibre could match the IFSS performance of glass fibre. The results of the pull out test show that the IFSS of PET – PP increase from 1.3MPa to 5.5MPa when increasing the MA content from 0% to 10% with maximum GF – PP IFSS reaching 10.2 MPa at 1.5% MA.

1. Introduction

Over the last decade the automotive industry has increasingly been using Polypropylene (PP) as a matrix material for fibre reinforced composites. This can be attributed to the material's attractive mechanical properties and low product cost ratio. Various fibres have been used to reinforce the PP matrix with Glass Fibre (GF) being one of the main reinforcement fibres. The automotive industry is investigating various routes to reduce energy consumption and environmental impact by improving the fuel efficiency of vehicles through weight reduction. One possible route is the substitution of GF reinforcement used in the PP composite with lighter, more flexible and more environmentally friendly fibres. Polyethylene Terephthalate (PET) fibre has been investigated as an alternative to GF. PET is a synthetic, aromatic and semi crystalline thermoplastic polymer which is currently being used in the manufacturing of plastic bottles[1, 2] . The polymer has numerous characteristics such as thermal stability, low density, resistance to the environment and recyclability that make it an attractive proposition. Recent research has shown that PP-PET composites have good impact properties [3]. Fibre reinforced composite performance is strongly influenced by the stress transfer capability of the fibre matrix interface. A critical requirement for a successful reinforcement is an excellent interfacial bond which will guarantee load transfer from the matrix to the fibre reinforcement. This paper investigates bond strength between PET fibre and PP containing different weight fractions of maleic anhydride (MA) by single fibre pull out. The results of PET-PP pull out are compared to GF-PP to investigate the means of PET fibre could match the IFSS performance of glass fibre.

2. Experimental

2.1 Materials

Polyethylene Terephthalate fibre with average diameter of 19µm, homopolymer polypropylene (579S MFI 47), and extrusion blended mixtures of various concentration PP + Maleic Anhydride Polypropylene (Polybond 3200) (MAPP) were obtained from SABIC. PP compatible glass fibre (GF) with an average diameter of 19 µm was sourced from Owens Corning Vetrotex.

2.2 Sample Preparation

The method employed in manufacturing single fibre pull out samples is illustrated in Figure 1.

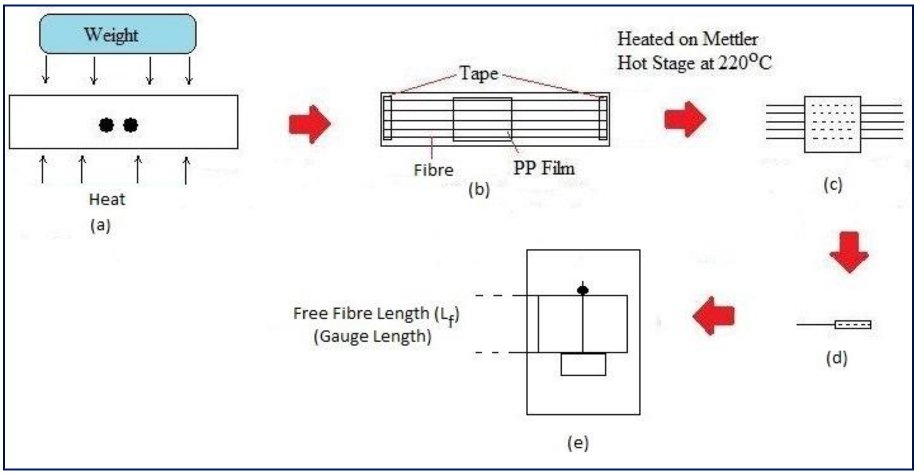

Figure 1: Pull Out Sample Preparation

PP pellets were heated on a hot stage between two glass slides at 220^{o}C and then a weight was applied creating a holding pressure on top of the slide sandwiching the pellets (Figure 1a). This created a film thickness between 0.15 – 0.2mm. Single fibres were then laid across a PP film on a glass slide with the fibre being held in place using tape. A second PP film was then carefully placed on top of the fibre and film ensuring both films were aligned accurately (Figure 1b). Before heat was applied to the film a glass cover slip was placed on top of the PP films to reduce oxidation during heating. The fibre and film (sample) were then placed in a Mettler FP 82 hot stage for four minutes at 220^{o}C to allow time for the fibre to be embedded in to the PP film [4]. The sample was then cut in half which created twice as many samples that had gone through the manufacturing process. Single fibre embedded in PP film was then separated and mounted on to card with superglue with a free fibre length of 5mm (Figure1 d and e). The diameter of each fibre tested was measured using the Nikon Epiphot inverted optical microscope and using Image analysis software, Image J.

2.3 Pull out Test

The pull out test was carried out at room temperature on Instron 3342 Tensile Test machine which was fitted with a device to carry out the pull out test. The device had two movable knife edges which were controlled by a pair of micrometer heads with a resolution of 1µm [5]. The knife edges were used to constrain the PP film with the distance between the blades being the diameter of fibre (Figure 2). A stereo microscope was used to assist the placement of the knife edges and to observe pull out test process. A load cell of 10N was used and the crosshead extension rate of 0.1mm/min was set for the test. Specimens with different embedded fibre length were tested and a minimum of 15 samples were tested for each MA weight fraction.

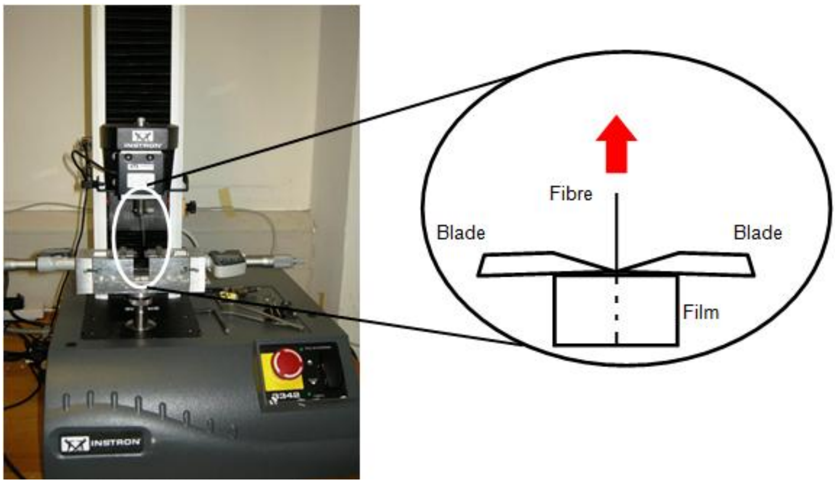

Figure 2: Pull Out Setup

3. Results and Discussion

3.1 Pull out Results

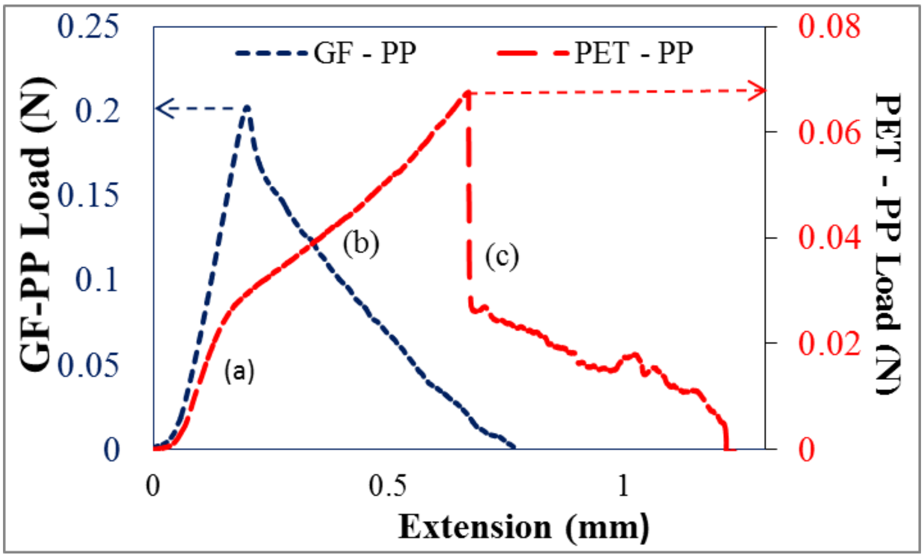

Figure 3: PET - PP and GF - PP Pull out Graph

Typical load versus extension graphs of PET-PP and GF-PP samples with a similar embedded area are presented in Figure 3. Comparing the two pull out graphs it can be seen that the maximum force required to debond the fibre from the matrix for GF is greater than PET's maximum force. Figure 3 also shows that the shapes of the two graphs are different. With GF the load increases to the maximum force where the fibre debonds from the matrix and then the force decreases gradually overcoming friction. Whereas to explain the PET – PP pull out graph it is necessary to split the graph into three stages.

a. The force increases due to the fibre being stretched up to a certain load. At this load the graph's gradient changes creating a 'kink' in the pull out plot. The cause of the 'kink' is discussed later in the paper.
b. The force is still increasing after the 'kink' until a maximum force is achieved. At this force the fibre debonds from the matrix.
c. The force suddenly drops to a lower load immediately after debond. The force then decreases gradually as the fibre is being pulled out of the matrix overcoming frictional forces.

The interfacial shear strength was calculated by using the Tyson – Kelly equation [4,6,7]:

$$\tau_{IFSS} = \frac{F_{\max}}{\pi D l_e} \qquad (1)$$

Where F_{max} is the maximum force from the pull out graph, D is the diameter of the fibre and l_e is the embedded length. The average IFSS (with 95% confidence) versus percentage of MA is graphed in Figure 4 for PET-PP and GF-PP. Figure 4 shows that increasing the MA percentage from 0 to 1.5% increases the GF IFSS considerably by 65% but has no significant effect on the IFSS of PET. The IFSS of PET-PP increases by approximately 70% when increasing from 1.5 to 5% MA but decreases by approximately 10% for GF-PP. Both fibres IFSS does not change between 5 – 10%MA. Comparing the interfacial strength of PET – PP and GF – PP it can be seen that the PET- PP IFSS are much lower than GF – PP IFSS. The IFSS of PET – PP at 5% MA is equivalent to 0.5% MA for GF – PP. The maximum IFSS that was achieved for PET was 5.6MPa and 10.2MPa for GF. It can be established from Figure 4 that MA improves the interfacial strength of both fibres but only up to a certain percentage of MA, 5% for PET and 1.5% for GF. The results show that a large amount of MA is needed for PET to achieve similar GF IFSS with a small amount of MA. Therefore it can be concluded that the interfacial bond between PET and PP must improve if the PET is to be used as an alternative to GF. One reason for the difference in IFSS could be due to the GF having an optimum sizing which helps with bond with PP whereas PET fibre has not.

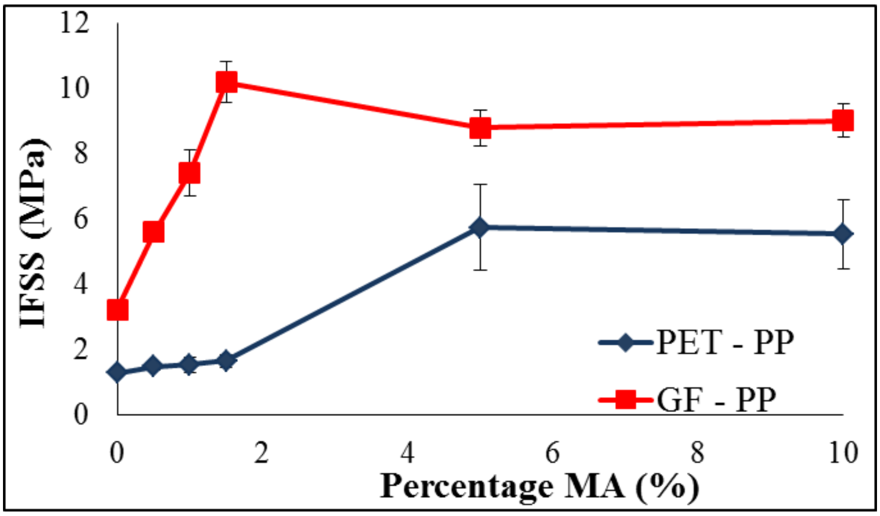

Figure 4: Interfacial Strength (IFSS) Versus Percentage Maleic Anhydride (MA)

3.2 'Kink' Investigation

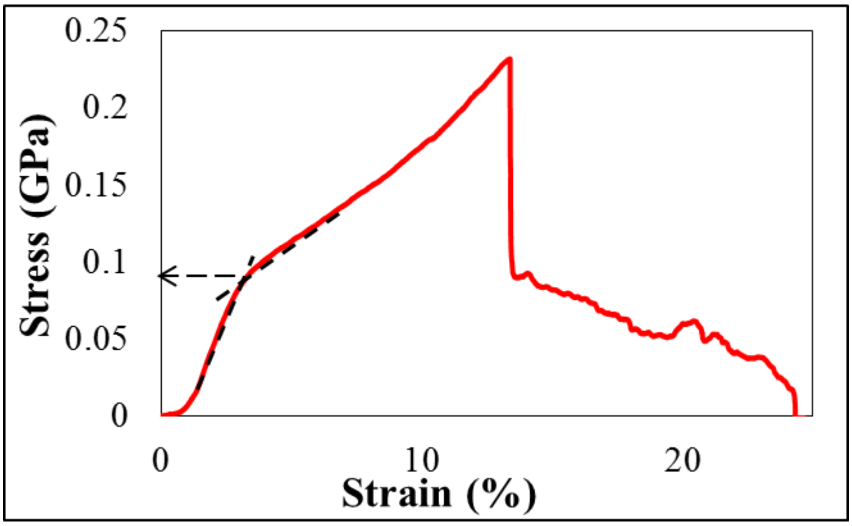

Figure 5: Stress versus Strain PET - PP Pull Out Graph

The PET-PP pull out graph was investigated further due to an unexplained change in slope ('kink') in the initial stage of the test. The 'kink' force for all of the pull out tests was determined by extending the gradient of the graph before and after the 'kink'. An example of calculating the 'kink' force is illustrated in stress against strain graph in Figure 5.

The 'kink' stress of all pull out test was plotted against the embedded area and can be seen in Figure 6. It was observed that the 'kink' occurred at approximately the same stress even with the percentage of MA increasing and different embedded areas. The stress at which the 'kink' occurs was between 0.06GPa and 0.1GPa.

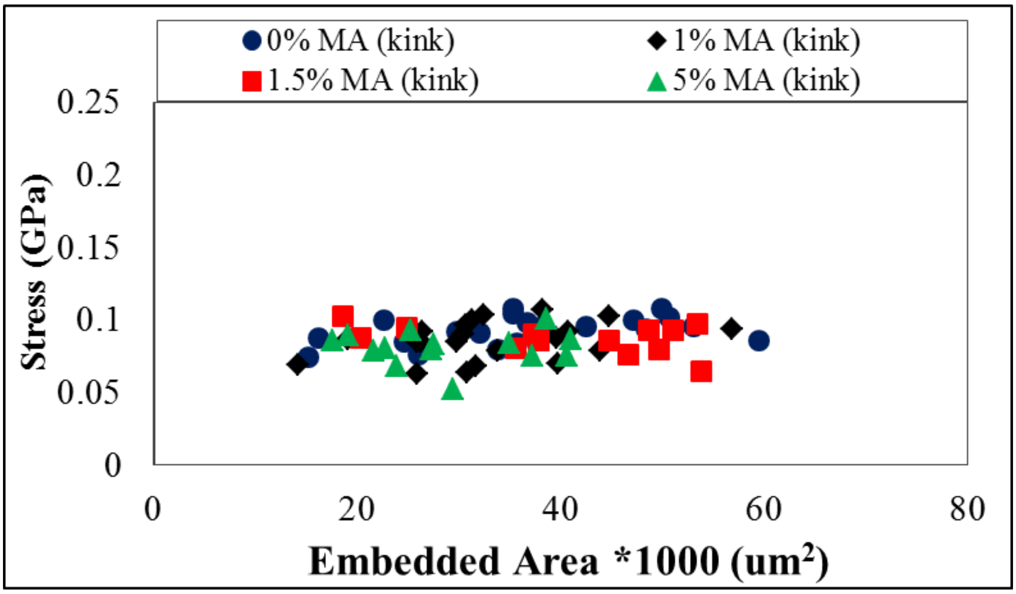

Figure 6: Maximum and kink force versus Embedded Area

A 'kink' phenomenon has been identified in the force – displacement results of IFSS tests by other researchers [6,7]. Mader et al noticed a 'kink' when undertaking pull out with GF-PP and described this as the fibre initially debonding from the matrix [6]. This is one possible explanation for what is happening but the system differs in one key way as the fibre that is being used is a polymer which is more ductile than GF. Another reason that could cause the change in slope in the pull out graph is the fibre. This reason was investigated further by carrying out tensile tests on PET fibre. Tensile tests were carried out on heat treated and non-heat treated PET fibre. The heat treated fibres went through the same manufacturing process for PET-PP pull out samples. Individual fibres were heated in the hot stage at 220^{o}C for four minutes and then removed to cool. The gauge lengths of the fibres were 5mm which was identical to the free fibre length for the pull out specimens. A minimum of ten tensile tests were carried out for the heat treated and non-heat treated fibres. The tensile test graphs of the two fibres were compared to the pull out graphs. To compare the pull out and tensile graphs a mutual start point of 0.01N was set therefore the extension/ strain was reset to 0 and an end point of 6% strain was decided for comparability reasons. These points were decided upon as they represented the range at which the 'kink' occurred. Figure 7 compares the average stress against strain of the pull out, heat treated and non-heat treated fibre tensile test.

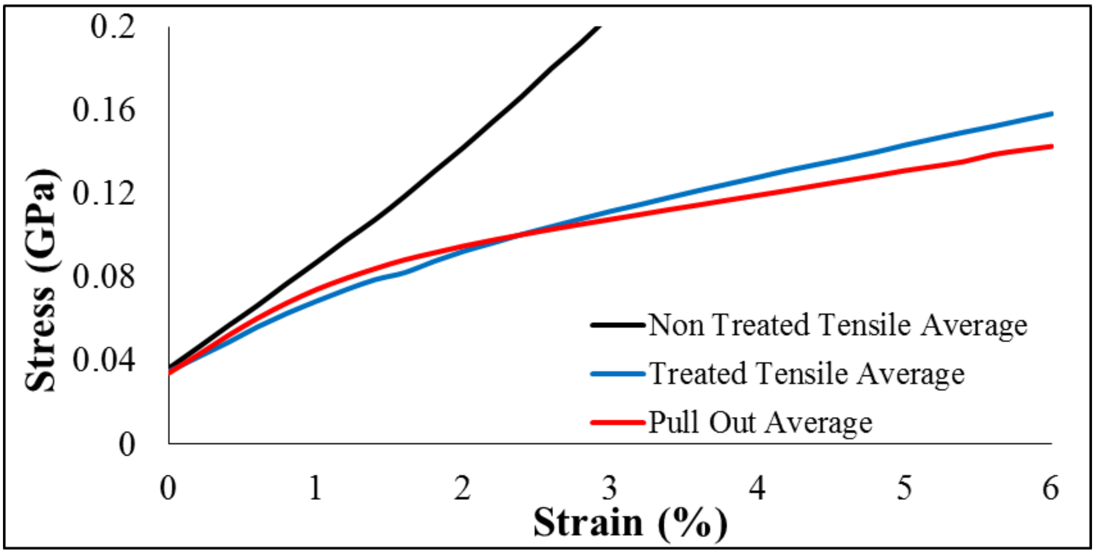

Figure 7: Average Stress Versus Strain graph of Pull out, Heat treated and non-heat treated fibre

Figure 7 shows that the stress strain curve of the heat treated fibre plotted overlays the pull out graph. It therefore seems very likely the fibre that is the causing the slope of the IFSS graph to change. Comparing the heat treated fibre and non-heat treated fibre stress strain curves it can be seen that heat treating the fibre changes the shape of the stress strain graph. This result has been identified by researchers such as Gupta et al and Cho et al [8–10]. Gupta carried out a series of experiments to investigate the effect of heat setting on the PET fibres' mechanical and structural properties [8, 9, 11, 12] In their studies they found that heat treating fibres with no mechanical constraint (free to relax) changed the fibre's structure considerably. Therefore the heat treatment on the 'free heated' fibre causes the amorphous and crystalline region of the fibre to be series and lowers the number of taut interlamellar ties. In the initial stage of the tensile test the tie molecules take all of the force. Due to the reduction of tie molecules, the stress on the tie molecules is high. This causes them to break at low load which leads to the fibre yielding. The stress is then applied to the molecules in the amorphous region where strain hardening may occur [8- 10]. From previous research that has been carried out on heat treated PET fibre and together with the results from Figure 7 it can be determined that the cause of the ‘kink’ is due to the fibre being heat treated at 220^{o}C in the manufacturing of samples. The ‘kink’ can be classified as the fibre’s yield point.

4. Conclusion

Single fibre pull out test were carried out on polyethylene terephthalate fibre and glass fibre embedded in polypropylene matrix containing various percentage of maleic anhydride. The graphs of pull out for both fibres were compared and it was found that a larger force is needed to debond GF from PP than the PET fibre. Further investigation in to the PET-PP pull out graph was required as a ‘kink’ appeared as the load was being applied to the system. It was discovered that the ‘kink’ in the PET-PP pull out graph was caused by the fibre being heat treated in the sample manufacturing process. Heat treating the fibre causes the fibre to yield at a low load therefore the ‘kink’ in the PET-PP is the yield point of the PET fibre. The IFSS results of the two fibres shows that by increasing the MA content the IFSS increased but only up to a certain MA percentage. The maximum IFSS for GF-PP was 10.2MPa at 1.5% MA and for PET-PP 5.5MPa at 5% MA. To conclude if PET fibre is to be considered in the automotive industry the interfacial strength will need to improve if to rival GF as reinforcement to PP.

5. References

[1] J. C. Viana, N. M. Alves, and J. F. Mano, “Morphology and mechanical properties of injection molded poly(ethylene terephthalate),” *Polymer Engineering and Science*, vol. 44, no. 12, pp. 2174-2184, Dec. 2004.

[2] R. Guzatto, M. Becker da Roza, E. L. Gasparotto Denardin, and D. Samios, “Dynamical, morphological and mechanical properties of poly(ethylene terephthalate) deformed by plane strain compression,” *Polymer Testing*, vol. 28, no. 1, pp. 24-29, Feb. 2009.

[3] C. Tufano and M. Soliman, “Polyethylene Terephthalate reinforced thermoplastic composites,” *ANTEC Conference*, 2010.

[4] L. Yang and J. L. Thomason, “Interface strength in glass fibre–polypropylene measured using the fibre pull-out and microbond methods,” *Composites Part A: Applied Science and Manufacturing*, vol. 41, no. 9, pp. 1077-1083, Sep. 2010.

[5] J. P. Craven, R. Cripps, and C. Viney, “Evaluating the silk/epoxy interface by means of the Microbond Test,” *Composites Part A: Applied Science and Manufacturing*, vol. 31, no. 7, pp. 653-660, Jul. 2000.

[6] E. Pisanova, S. Zhandarov, and E. Mader, “How can adhesion be determined from micromechanical tests ?,”*Composites Part A: Applied Science and Manufacturing*, vol. 32, pp. 425-434, 2001.

[7] A. A. Apostolov, M. Evstatiev, Z. Denchev, K. Friedrich, and S. Fakirov, “Effect of composition on transcrystallization with reorientation of polypropylene in drawn PET/PP blend,” *Journal of Materials Science*, vol. 42, no. 4, pp. 1245-1250, Jan. 2007.

[8] V. B. Gupta and S. Kumar, “The Effect of Heat Setting on the Structure and Mechanical Properties of Poly (ethy1ene Terephthalate) Fiber . IV . Tensile Properties Other Than Modulus and Their Dependence on Structure,” *Polymer*, vol. 26, pp. 1897-1905, 1981.

[9] V. B. Gupta and S. Kumar, “The Effect of Heat Setting on the Structure and Mechanical Properties of Poly (ethy1ene Terephthalate) Fiber . 111 . Anelastic Properties and Their Dependence on Structure,” *Polymer*, vol. 26, pp. 1885-1895, 1981.

[10] D. Hwan Cho, W.-R. Yu, J. H. Youk, and J. H. Yoo, "POLYMER Formation of micro-crystals in poly (ethylene terephthalate) fiber by a short heat treatment and their influence on the mechanical properties," *European Polymer Journal*, vol. 43, pp. 3562-3572, 2007.

[11] V. B. Gupta and S. Kumar, "The Effect of Heat Setting on the Structure and Mechanical Properties of Poly (ethy1ene Terephthalate) Fiber . I . Structural Changes," *Polymer*, vol. 26, pp. 1865-1876, 1981.

[12] V. B. Gupta and S. Kumar, "The Effect of Heat Setting on the Structure and Mechanical Properties of Poly (ethy1ene Terephthalate) Fiber . 11 . The Elastic Modulus and Its Dependence on Structure," *Polymer*, vol. 26, pp. 1877-1884, 1981.