Mechanical Properties of Low-Density Polyethylene/Recycled Poly(ethylene Terephthalate) (LDPE/r-PET) Microfibrillar Composite

by

Nabila Farhana binti Abdul Khalil

Dissertation submitted in partial fulfilment of

the requirements for the

Bachelor of Engineering (Hons)

(Mechanical Engineering)

SEPTEMBER 2011

Universiti Teknologi PETRONAS
Bandar Seri Iskandar
31750 Tronoh
Perak Darul Ridzuan

CERTIFICATION OF APPROVAL

Mechanical Properties of Low-density Polyethylene/Recycled Poly(ethylene terephthalate) (LDPE/r-PET) Microfibrillar Composite

by

Nabila Farhana binti Abdul Khalil

A project dissertation submitted to the
Mechanical Engineering Programme
in partial fulfilment of the requirements for the
Bachelor of Engineering (Hons)
(MECHANICAL ENGINEERING)

Approved by,

_____

(Dr. Mohamad Zaki bin Abdullah)

UNIVERSITI TEKNOLOGI PETRONAS

TRONOH, PERAK

September 2011

CERTIFICATION OF ORIGINALITY

This is to certify that I am responsible for the work submitted in this project, that the original work is my own except as specified in the references and acknowledgements, and that the original work contained herein have not been undertaken or done by unspecified sources or persons.

_____

NABILA FARHANA BINTI ABDUL KHALIL

ABSTRACT

Blending can help improving mechanical properties of materials. It is a cheaper way to tailor mechanical properties of the material instead of polymerization. But, blending of immiscible polymers will result in low mechanical properties as there are no connections between the materials and the sphere shape of the polymer. So, microfibrillar composite (MFC) is produced by drawing process and act as reinforcement in the material. While compatibilizer helps in improving interfacial adhesion between the microfibrillar and matrix. So, with aid of reinforcement and compatibilizer, mechanical properties of the blend can be improved. Aim of this study is to see the effects of microfibrillar towards the mechanical properties of LDPE/r-PET composite and to study the effects of compatibilizer towards the mechanical properties of LDPE/r-PET composite. In this study, LDPE is used as the matrix and recycled PET (r-PET) was employed as the reinforcement. LDPE and r-PET is mixed using twin screw extruder and was drawn to produce the microfibrils. The blend is then compression moulded to produce specimens for mechanical tests. Tensile, flexural and impact tests are done base on ASTM D368, D790 and D256 respectively to study mechanical properties of the composite. Microfibrillar is proven to improve tensile strength of drawn LDPE/r-PET by 6% when compared with undrawn LDPE/r-PET and improve flexural strength by 13%. When comparing with pure LDPE, tensile strength of drawn LDPE/r-PET increased by 10% and flexural strength increased by 126%. Coupling agent managed to improve tensile strength of the undrawn LDPE/r-PET/Coupling agent composite by 12% compared than undrawn LDPE/r-PET and improve flexural strength by 21%. Compared to pure LDPE, tensile strength of drawn LDPE/r-PET increased by 18% while flexural strength increased by 155%. Impact strength of drawn LDPE/r-PET improved by 10% compared than undrawn LDPE/r-PET but it is reduced by 95% when compared to pure LDPE. For undrawn LDPE/r-PET/coupling agent specimen, impact strength improved by 9% compared to undrawn LDPE/r-PET but when compared to pure LDPE, impact strength value is decreased by 96%.

ACKNOWLEDGEMENT

First and foremost, I would like to express my utmost gratitude to my supervisor, Dr. Mohamad Zaki bin Abdullah for all his guidance, motivation and encouragement throughout completion of this project. The supervision and support that he gave truly help the progression and smoothness of this project.

My heartiest appreciation to PRSB staffs - Mr. Yusof Said, Mr. M Rossi M Bahari, Mr. Navin Sharma and Mr. Shamsul Farid for giving me permission to use equipments in PRSB to complete my project and for the guidance given throughout my stay at PRSB. Thanks you for sharing vast knowledge on research as well as polymers that has been a real privilege.

Special thank to Mr. Syairul Harun, UTP Material Lab Technician, who are willing to work over the weekend to assist me with the lab equipments and not to forget, Mr Irwan. I would also like to express my heartfelt appreciation to all fellow friends who have unconditionally lent a helping hand throughout my project.

Finally, words alone cannot express the thanks I owe to my family especially my parents for their never ending supports and encouragement that contribute towards the completion of this project.

TABLE OF CONTENTS

LIST OF FIGURES

LIST OF TABLES

CHAPTER 1

INTRODUCTION

1.1 Background of Study

Polymer is pure material which results from polymerisation process and is usually taken as the family name for materials that have long chain-line molecules [1]. Polymer and polymer composites have several advantages including light weight, good processability, chemical stability in any atmosphere condition and many more. Thus, they have been widely used in many areas like engineering, electronics and transportation [2].

Polymer blending is a common processing technique used to create new materials with desired qualities are either retained or usually enhanced which inherent to one or the other blend constituents [3]-[4]. Blending is a versatile and economical method to manufacture products to satisfy complex performance demand [5]-[7]. Blending of polyolefins and poly(ethylene terephthalate) (PET) has been the area of interest since it can help improving mechanical properties of the relatively weak partner [4], [6]. Nowadays, composite materials have replaced the traditional ones in variety of applications, mainly because they are light in weight but having enhanced mechanical properties [8].

Morphology and properties of immiscible or partially immiscible blends can be improved with incorporation of compatibilizers [3], [6]-[7]. Compatibilizer helps in enhancing interfacial adhesion between the immiscible blend components and improving properties of the material.

As plastic is dominating the municipal solid waste fraction in the world, recycling plastics can help reducing the resources needed in manufacturing, conserve energy in production and shipping and also minimizing the overall impact on the environment over life-cycle of the product. Solid household waste is made up of a

mixture of largely polyolefin-based resin including poly(ethylene terephthalate) (PET) [6], [9]-[10].

1.2 Problem Statement

Polymer blending can help improving the mechanical properties of materials with relatively low cost instead of polymerization. But, blending of two immiscible polymers will result in poor mechanical properties due to shape of the dispersed phase and no connection between the materials. So microfibrils are formed using drawing technique that act as the reinforcement to help in improving mechanical properties of the material. Meanwhile, compatibilizer is used to provide better interfacial connection between the materials.

Meanwhile, plastics are used to create many household items as well as for packaging. As plastics are non-biodegradable material, it will take hundred years to decompose. So recycling or reusing of plastics can help saving the environment.

1.3 Objectives

The objectives of this project are:

a) To study the effects of microfibrillar on the mechanical properties of LDPE/r-PET composite.
b) To study the effects of compatibilizer on the mechanical properties of LDPE/r-PET composite.

1.4 Scope of Study

This project is mainly focusing on microfibrillar composite (MFC) which involves LDPE as the matrix and r-PET as the reinforcement with the ratio of 70/30. r-PET microfibrillar is formed by drawing. LDPE/r-PET blend is then compression moulded to produce specimens for testing purpose. Tensile, flexural and impact tests are conducted to determine mechanical properties of the MFC according to ASTM D638 [11], D790 [12] and D256 [13] respectively. Compatibilizer is also added to

improve interfacial adhesion between the matrix and reinforcement and finally improving the mechanical properties.

1.5 Significance of Study

Microfibrillar composite can help enhancing mechanical properties of the matrix as well as the reinforcement material. It is also a cheaper way to tailor the properties of a product as compared with polymerization. Apart from that, recycled PET is used to create the blend which can help saving the environment by minimizing the usage of virgin plastics like PET.

1.6 Feasibility of Project

This project will require some experimental works in fabricating the specimen and to conduct mechanical tests to determine the specimen's tensile, flexural and impact strength. So, progress of this project will depends on condition and availability of the machines which are grinder, extruder, compression moulding machine, Universal Testing Machine (UTM), impact tester and Scanning Electron Microscope (SEM).

This project can be done within the allocated time given that everything goes fine as planned. All of the objectives can be achieved if the procedures are followed closely. As this is a small-scale project, not much material is needed and most of the materials used are already available in UTP Material lab. As most of the experiments are done using UTP's lab equipment, so, the allocated RM500 budget will be sufficient enough to complete this project.

CHAPTER 2

THEORY AND LITERATURE REVIEW

2.1 Overview of Microfibrillar Composite (MFC)

A fibril can be defined as a structural entity with material properties that are biased predominantly along a linear dimension or symmetry axis [14]. While microfibrillar composite (MFC) is referring to materials in which the reinforcing elements are polymer microfibrils [8], [15]-[16]. It comprises of two immiscible thermoplastic polymers which have distinct difference in their melting temperatures (at least 40°C) [3]-[4], [8], [14], [17]-[23]. The constituent polymers must have adequate drawability to allow formation of the microfibrils to occur [3]-[4]. This type of material provides a way to tailor the properties of a product for desired application [15]-[16].

MFCs are found to have significantly enhanced mechanical properties compared than the conventional blends as well as the virgin compatibilized blends [6], [14], [19]-[21], [24]. However, the properties and applications are depending on the morphology of the created polymer fibrils and their thermal stability [19]. Good mechanical properties in a composite material can be achieved if there is high aspect ratio as well as good adhesion between the matrix and the reinforcing phase [21]-[25]. Adhesion between the reinforcing element and the matrix can be improved using coupling agent or compatibilizer [22], [25]. In presence of compatibilizer, morphology of the resulting blends shows good dispersion of fine minor phase [22].

The combination of polyolefins such as polypropylene (PP) and polyethylene (PE) with poly(ethylene terephthalate) (PET) has been tried by many researchers. PET is preferred because of its inherent fibre forming capability while PE or PP is chosen because of economic and processability [6].

Since the microfibrils are not available in separate material, it has to be created [3], [14], [22]-[25]. It is formed by melt-blending of two immiscible thermoplastic polymers, followed by hot or cold drawing with speed difference between rollers must be at least 5 times [14] and annealing (thermal treatment) of the drawn blend at constant strain and at $T_1 < T < T_2$ where T_1 and T_2 are the melting temperatures of the two components [4], [6]-[8], [14]-[17], [19]-[27]. Both of the constituent polymers must be able to be processed at a single temperature without the onset of degradation in either polymer [3]. During the thermal treatment, the processing temperature must be not too close with the melting temperature of the microfibrils. If not, they will melt again and return back to their spherical shape [14], [20]. The microfibrils are created after blending of the two materials which upon drawing, the blends are oriented and microfibrils are formed in discontinuous form [8], [15], [18].

Annealing is done for perfection of the structure. Temperature and duration of annealing have been shown significantly to affect the structure and properties of the blend. If the annealing temperature (T_a) is set below the melting point of both components, the microfibrillar structure imparted by drawing is preserved and furthermore improved as a result of physical processes such as additional crystallization, minimization of defects in crystalline regions and relaxation of residual stresses in amorphous region. If the annealing temperature (T_a) is set between the melting point of both components, isotropization of the low melting component takes place to form an isotropic matrix. While, higher melting temperature preserve their oriental and morphological characteristics [28].

Morphology of dispersed phase in the blends can greatly affects the physical and mechanical properties of the composite. For example, well-dispersed and fine spheres in the matrix can help increasing impact strength of a material, while sheet-like dispersed morphology has the ability to enhance barrier properties of a film, and microfibrils can increase the uniaxial strength greatly [16].

Nanoparticle reinforcement in polymeric matrix material can lead to significant property improvements or in some cases, they may worsen the properties because the nanoparticles has strong tendency to agglomerate in polymeric matrices [8]. According to Jayanarayanan *et al.* [24], a very high draw ratio during cold drawing of PP/PET will lead to the attrition of the microfibrils in microfibrillar blends (MFB) and brings down the mechanical properties of the MFCs. Draw ratio

can be done only up to 8 times. Beyond that, breakage of fibrils was observed during stretching and results in low mechanical properties value [23].

MFCs can be used as a packaging material as it is easy to manufacture and it is suitable for many common polymer resins [3].

2.2 The Effects of Blending Ratio towards Mechanical Properties of the Blends

Blending ratio of matrix and the reinforcement material gives effects on mechanical properties of the blend. Many research have been done on MFCs and 70/30 ratio of PE/PET is proven as the most effective one. A summary of blend ratio and mechanical properties of the blend is shown in Table 2.1.

Table 2.1: The effects of blending ratio on mechanical properties of the blends [29].

Author(s)	**Material**	**Ratio**	**Tensile Strength (MPa)**	**Flexural Strength (MPa)**	**Impact Energy (kJ/m^2)**
Evstatiev *et al.*	LDPE/PET	100/0	8.8		53
		70/30	18		66
		50/50	27		75
Fakirov *et al.*	LDPE/PET	100/0	8		
		70/30	17		
		50/50	26		
Li *et al.*	PE/PET	95/5	22.0		
		90/10	25.2		
		85/15	31.3		
		80/20	32.8		
		75/25	35.9		
		70/30	29.2		
		65/35	24.5		
Li *et al.*	PE/PET	90/10	26		
		85/15	31		
		80/20	33		

		75/25	36		
Li *et al.*	HDE/PET	100/0	20.5		
		85/15	23-31		
		0/100	78		
Li *et al.*	PE/PET	100/0	20.5		
		95/5	21.1		
		90/10	22.0		
		85/15	22.9		
		80/20	23.7		

2.3 The Effects of Compatibilizer

Compatibilizer or coupling agent, as the name suggest, is used to improve compatibility between immiscible polymers. In MFC, compatibilizer will improve interfacial adhesion between matrix polymer and reinforcement polymer [22]. Taepaiboon *et al*. has proved that compatibilizer helps enhancing tensile, flexural and impact properties of the iPP/r-PET blends. PP-*g*-MA compatibilizer is used in the experiment and the most efficient ratio of iPP/r-PET/PP-*g*-MA blend is 70/30/5.

Ballauri *et al.* [30] has used Kraton as the compatibilizer for PET/PP blends. Adding 5% of the compatibilizer increased impact strength of the PET/PP blend by 4 times and increased elongation at break of the blend by 10 times compared to uncompatibilized blend.

2.4 Extrusion

Extrusion can be done using either single screw extruder or twin screw extruder. In extrusion, plastic in the form of pellets or powder is fed into a heated cylinder while rotating screw homogenise it and squeeze it through a die [31].

Single screw extruder is intended for thermoplastic materials in pellet or powder form [32]. The pellets are poured into the hopper, which delivers them to a screw [1]. The diameter of the screw root progressively increases along its axis in the direction of die. This accommodates the densification associated with the melting

processes wherein spaces between the particulates disappear. In the region where the polymer has melted, the extruder acts as a drag flow pump and pushes the melt into the die entrance.

While twin screw extruder, is mainly used for producing polyvinyl chloride profiles and pipes, as well as for polyolefin processing. There are various types of twin screw extruder. The screws may rotate in the same direction (co-rotating), in opposite direction (counter rotating) or the screws may be separated, tangential, intermeshing or self wiping [32].

2.5 Hot/Cold Drawing

Drawing is a process where polymer fibres are strengthened by elongation [33]. For hot drawing, device with three pinching rolls is used to form the microfibrils. The rolls temperature is kept at about 40°C by adjusting the tap water flow rate in the cooling pipe. Speed of the rolls can be adjusted to obtain different stretching ratio.

While for cold drawing, the material is drawn in dry hot air with temperature higher than glass transition temperature of PET. Speed of the rolls can be adjusted to achieve desired draw ratio. Cold drawing is primarily used in manufacturing plastic fibres.

2.6 Compression Moulding

Compression moulding is one of the oldest processes. It has been used by man thousand years ago to clay or natural resins [34]. Compression moulding involves the pressing (squeezing) of a deformable material charged between the two halves of heated mould and its transformation into a solid product under the effect of elevated temperature and pressure given [31].

A pre-weighted plastic is placed in the lower half of a heated mould and the upper half is the forced down. This causes the material to take the shape of the mould. The application of heat and pressure accelerates the polymerisation of the plastic and once the crosslinking is completed, the article is solid and may be ejected while still very hot [1].

2.7 Materials

2.7.1 Low Density Polyethylene (LDPE)

LDPE is so named because it contains substantial concentrations of branches that hinder crystallization process, resulting in low densities [36]. It is a semi-rigid, translucent material and was the first of the polyethylenes to be developed. It is mainly used in high-clarity products which includes produce bags, bakery film and textile overwrap. It also includes durable products such as power cables coating and toys [1], [31].

It qualities includes toughness, flexibility, resistance to chemical and weather and low water absorption [1], [36]. It is easily produced by most methods and has a low cost. It is a corrosion-resistant, low-density extruded material that provides low moisture permeability. LDPE has a fairly low working temperature, soft surface and low tensile strength. It is also lightweight and formable, has high impact resistance and excellent electrical properties [31].

Disadvantages of LDPE are it is low in strength, stiffness and maximum operating temperature. It also has poor UV resistance, flammability and susceptible to environmental stress cracking [31].

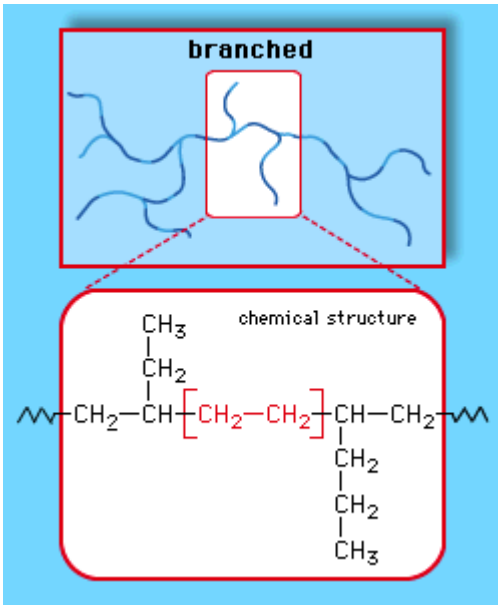

Figure 2.1: The branched form of polyethylene, which is called low-density polyethylene (LDPE) [35].

2.7.2 Poly(ethylene terephthalate) (PET)

PET is thermoplastic polymer widely in the area of packaging where optical clarity if the main concern [22]. Applications of PET also include textile fibres (sailcloth), films, and other moulded materials [7]. The stiffness of PET fibres makes them highly resistant to deformation, so that they are excellent in impact resistance. PET is hard, stiff, strong, dimensionally stable material that absorbs very little water. It is normally used in making plastic bottle.

One of the main reasons for the wide spread use of PET is their possibility of producing a number of different grades over a broad range of molecular weights in a single multiproduct polymerization plant. Chemical structure of PET is shown in Figure 2.2.

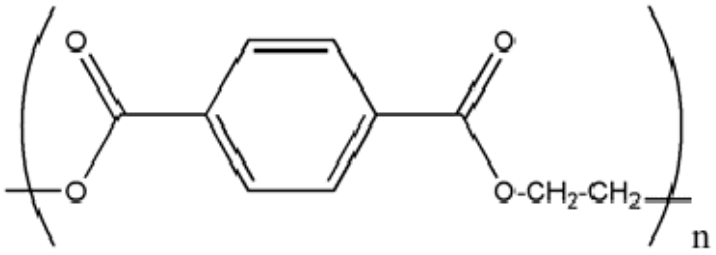

Figure 2.2: Poly(ethylene terephthalate) chemical structure [7].

Recycling of PET is quite common in the industry. Recycled PET (r-PET) has been used as a component in the production of parts and textile for automobiles and as a cost reduction ingredient for food containers [22], [36].

r-PET undergoes a reduction in intrinsic viscosity or molar mass when recycled in a normal extrusion system because of thermal and hydrolytic degradation. This results in significantly lower mechanical properties especially tensile [9].

CHAPTER 3

METHODOLOGY

3.1 Research Methodology

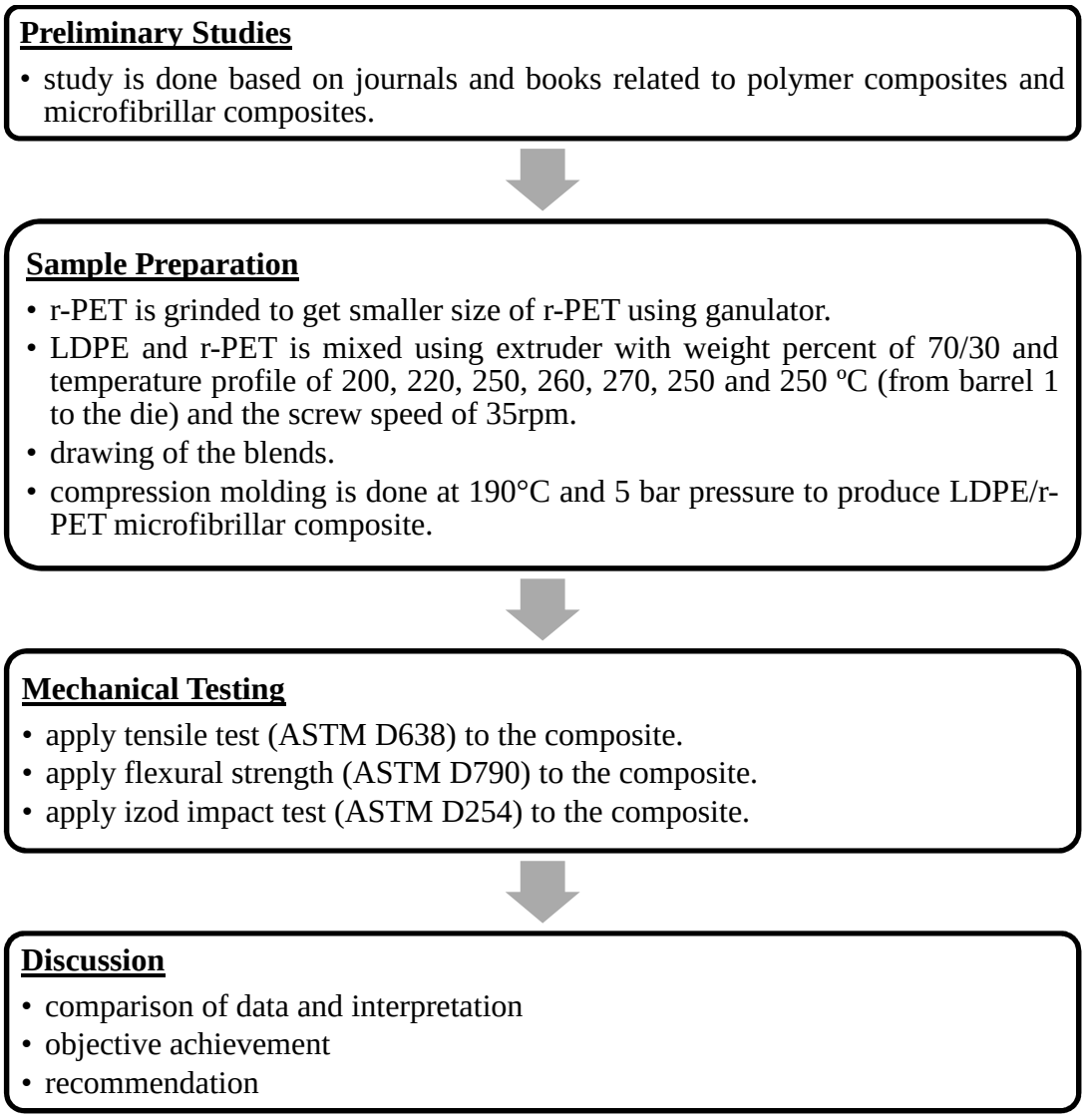

Figure 3.1: Research methodology.

Figure 3.1 summarizes the methodology of this study which includes preliminary studies, sample preparations, mechanical tests and discussion.

3.2 Experimental Works

3.2.1 Sample Preparation

Materials used in this project are:

a) *Low density polyethylene (LDPE):* TITANLENE LDI300YY from Titan Chemicals in granule form with melting temperature of 160 - 240ºC, tensile strength of 8 - 10 MPa, tensile modulus of 0.2 – 0.3 GPa, flexural modulus of 0.2 – 0.3 GPa and density of 0.000920 g/mm^3 as provided by manufacturer.

Figure 3.2: Raw LDPE granule.

b) *Recycled Poly(ethylene terephthalate) (r-PET):* recycled PET from drinking bottles (bottle grade) with melting temperature of 254 – 256ºC, tensile strength of 55 – 75 MPa, flexural strength of 80 – 120 MPa, flexural modulus of 2.3 – 3.0 GPa and density of 1.38 – 1.40 g/mm^3.

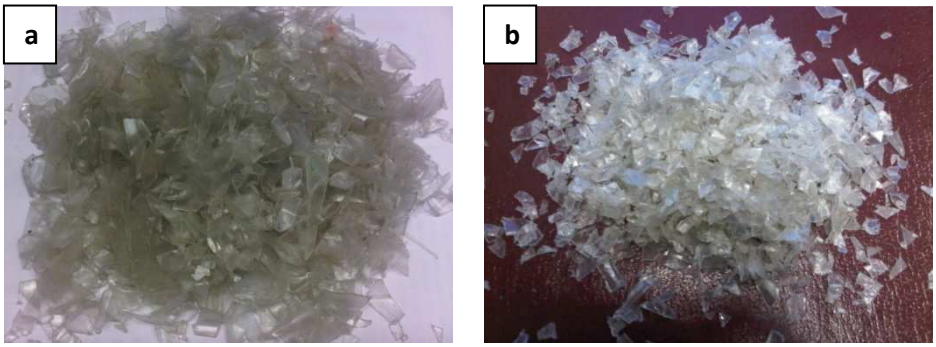

Figure 3.3: Raw r-PET flakes: (a) before grinding (b) grinded.

c) *Anhydride modified polypropylene (coupling agent):* Fusabond® P613 from DuPont™ in granule form with melting point of 162ºC, density of 0.903 g/cm^{3}and melt flow rate of 49 g/10 min.

Figure 3.4: The coupling agent granules.

To prepare the samples, r-PET from drinking water bottles are cut into small flakes. Then they are grinded in a granulator to get smaller size of r-PET flakes to ease the blending process. Five samples are made which consist of neat LDPE, LDPE/r-PET with ratio of 70/30, LDPE/r-PET (drawn) with ratio of 70/30, LDPE/r-PET/coupling agent with 70/30/5 ratio and LDPE/r-PET/coupling agent (drawn) with 70/30/5 ratio. They were dry-mixed first before being melt-mixed using Brabender Twin Screw Extruder with increasing temperature profile of 200, 220, 250, 260, 270, 250 and 250 ºC (from barrel 1 to the die) and the screw speed of 35rpm. Diameter of the die opening was 2 mm.

For the drawn mixture, drawing was done after the mixture has cooled down a bit (above T_g of PET to allow the molecules move freely during realignment) after

being extruded. Drawing was done manually where the extruded mixture is drawn 4 times from its original lengths.

Table 3.1: Composition of the samples.

Sample	Weight Ratio
Pure LDPE	100
LDPE/r-PET	70/30
LDPE/r-PET (drawn)	70/30
LDPE/r-PET/coupling agent	70/30/5
LDPE/r-PET/coupling agent (drawn)	70/30/5

Figure 3.5: Extruded LDPE/r-PET blend.

3.2.2 Moulding

The prepared sample is compression moulded with temperature of 190°C and 50 bar pressure. For the compression moulding process, the preheating time is 1 minute 11 second, moulding time is 5 minutes 28 second and cooling time is 5 minutes 28 second. The sample is cooled under 50 bar pressure.

Figure 3.6: Compression moulding machine used in preparing the specimen.

The moulded sample is then cut into dog-bone shape for tensile test using pressure cutter.

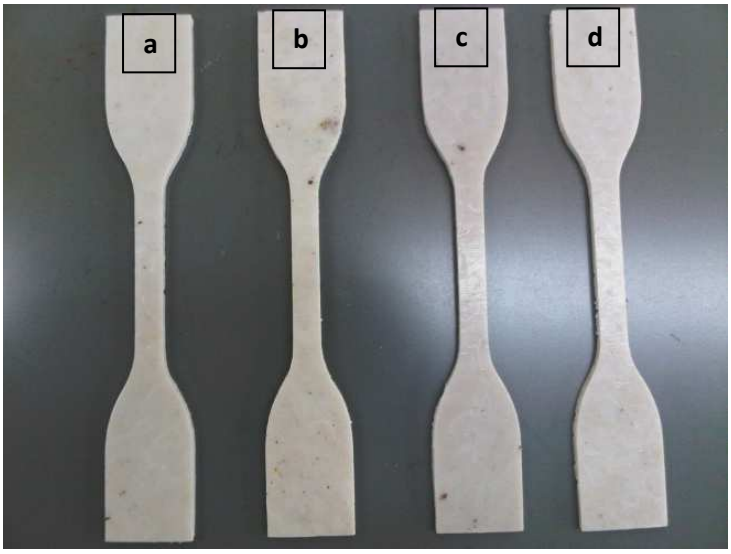

Figure 3.7: Samples for tensile test: (a) undrawn LDPE/r-PET (b) drawn LDPE/r-PET (c) undrawn LDPE/r-PET/coupling agent (d) drawn LDPE/r-PET/coupling agent.

Figure 3.8: Pressure cutter.

3.2.3 Mechanical Property Tests

Tensile, flexural and impact test is done base on standards ASTM D638, ASTM D790 and ASTM D256 respectively. Test should be done on at least five (5) specimens to get accurate result.

Table 3.2: Standards used in mechanical property tests.

Test	Standard
Tensile	ASTM D368
Flexural	ASTM D790
Impact (Izod)	ASTM D256

Tensile Test

Tensile test is done using Universal Testing Machine (UTM) in Petronas Research Sdn. Bhd. (PRSB). Samples were cut into Type IV dog-bone shape for the testing purpose. Type IV is used for specimen with thickness less than 4 mm. The cross-head speed was 50 mm/min and load cell used is 5 kN.

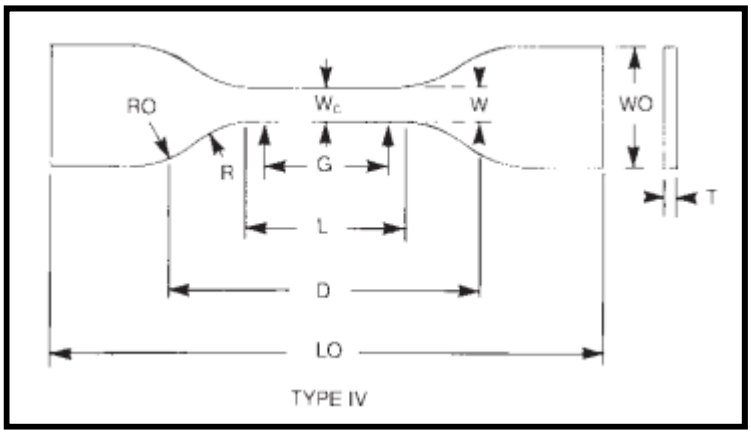

Figure 3.9: Type IV specimen.

Table 3.3: Type IV specimen dimension**.**

Dimensions	**Size (mm)**
W – width of narrow section	6
L – length of narrow section	33
WO – width overall	19
LO – length overall	115
G – Gage length	25
D – distance between grips	65
R – radius of fillet	14
RO – outer radius	25
T – thickness	4 or under

Figure 3.10: Universal Testing Machine used for tensile test.

Flexural Test

Flexural test is done using Universal Testing Machine (UTM) in Universiti Teknologi PETRONAS (UTP). Samples were cut according to the standard.

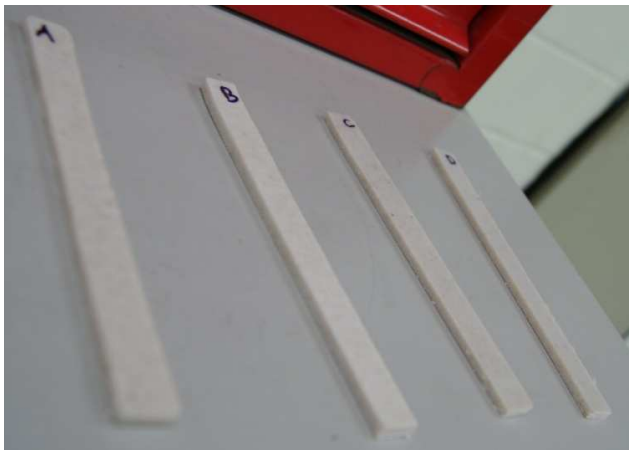

Figure 3.11: Samples for flexural test.

Impact Test

Impact test is done using impact tester in Petronas Research Sdn. Bhd. (PRSB).

Figure 3.12: Impact tester.

Sample was cut according to the standard and notch of 2.54 mm is made.

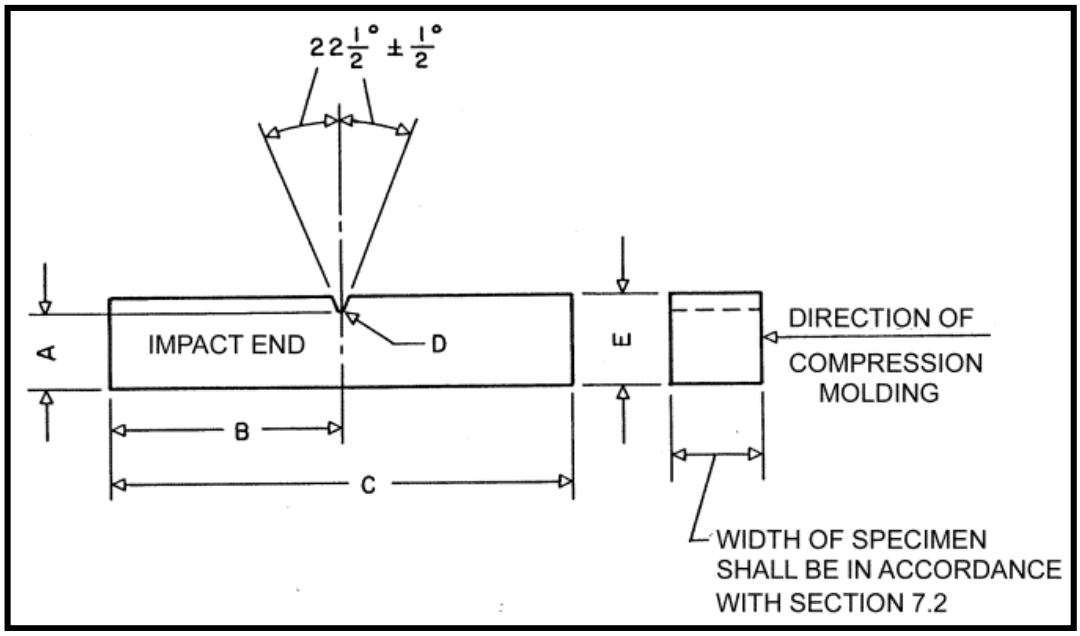

Figure 3.13: Izod impact test specimen specifications.

Table 3.4: Izod impact test specimen dimension.

Dimensions	Size (mm)
A	10.16 ± 0.05
B	31.8 ± 1.0
C	63.5 ± 2.0
D	0.25R ± 0.05
E	12.70 ± 0.20

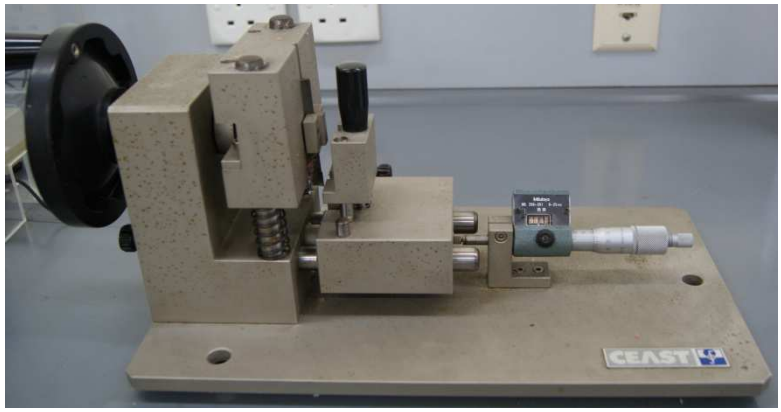

Figure 3.14: Tool to make notch.

3.3 Project Activities

Table 3.5: Project activities.

Activities	Starting Month	Finishing Month
Preliminary studies	20 June 2011	31 July 2011
Purchasing of materials needed	1 August 2011	20 August 2011
Extrusion and drawing of the mixed materials.	21 August 2011	30 September 2011
Compression moulding of the material and preparation for testing.	1 October 2011	31 October 2011
Applying tensile, flexural and impact tests to the material; and microscopic scanning	1 November 2011	30 November 2011
Report documentation	1 December 2011	15 January 2012

3.4 Gantt Chart and Key Milestone

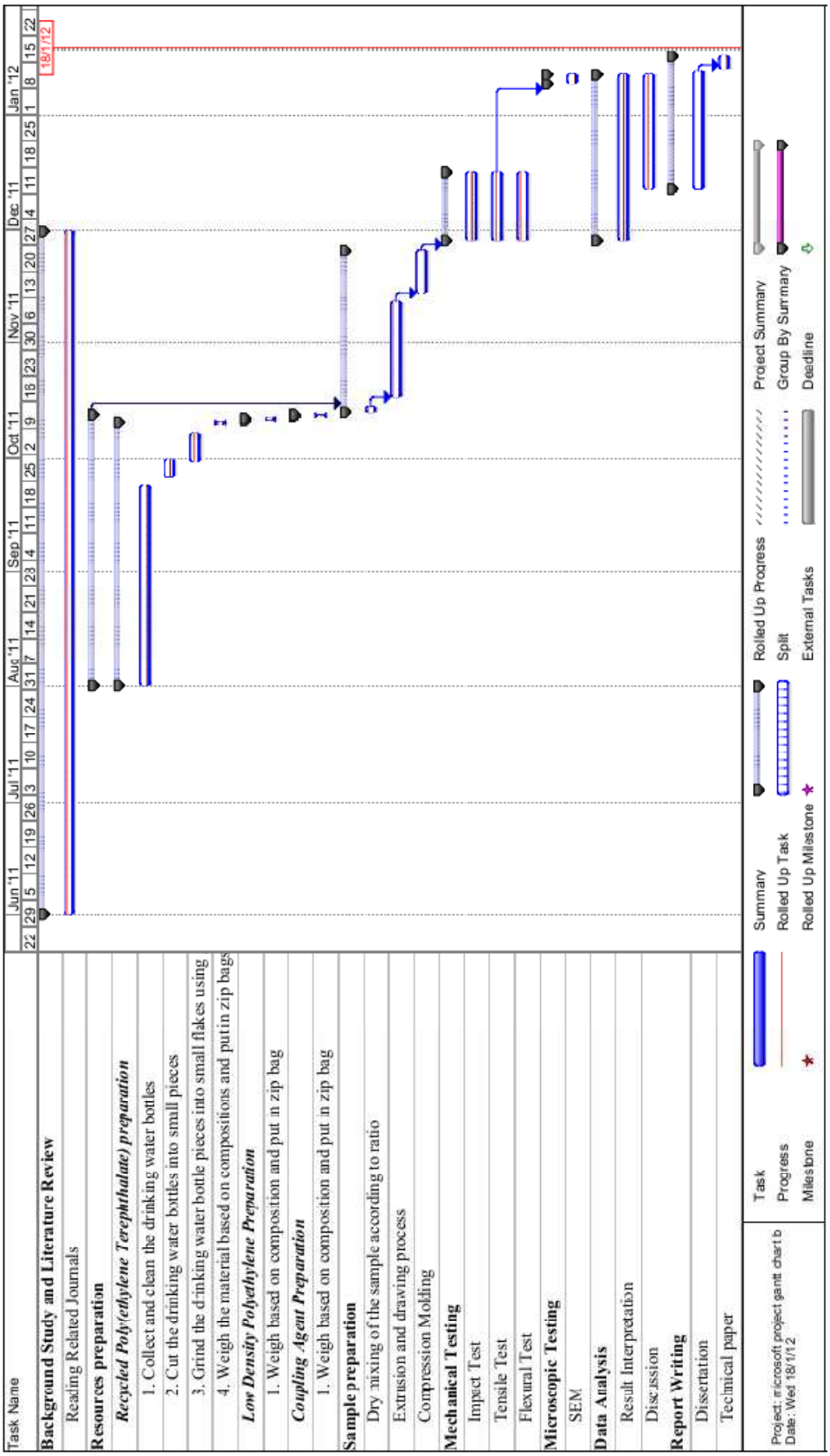

Figure 3.15: Gantt chart and key milestone of the project.

3.5 Tools and Equipments

Low speed granulator model SG-21P is used to grind the r-PET bottles to smaller size of r-PET flakes. Then, Leistritz twin screw extruder model Mio27/6l-32D is used for LDPE and r-PET mixing purposes. Pallletizer machine model C.F SCHEER is used to palletize the extruded LDPE and r-PET mixture before going through compression moulding process. Then, compression moulding is done using compression moulding machine at PETRONAS Research Sdn. Bhd. (PRSB). For testing purposes, Universal Testing Machine (UTM) at PRSB is used for tensile test.

Then, three-point bending method is used in determining flexural strength of the specimen. Izod impact test is done to determine the composite's impact strength. Scanning Electron Microscope (SEM) is used to examine morphology of the blends.

CHAPTER 4

RESULT AND DISCUSSION

4.1 Tensile Strength

Tensile test has been done to the specimens using Universal Testing Machine (UTM). Result of the test is shown in Table 4.1.

Table 4.1: Tensile strength of the specimens.

Sample	Tensile Strength (MPa)				
	Pure LDPE	Undrawn LDPE/r-PET	Drawn LDPE/r-PET	Undrawn LDPE/r-PET/ Coupling agent	Drawn LDPE/r-PET/ Coupling agent
1	6.58	7.57	8.19	7.89	9.04
2	8.28	8.15	8.64	8.32	8.58
3	6.95	7.76	8.43	9.88	11.03
4	8.35	8.13	8.33	9.69	11.19
5	7.81	7.93	8.21	8.20	9.48
Avg	**7.60**	**7.91**	**8.36**	**8.79**	**9.87**
Std	**0.80**	**0.25**	**0.18**	**0.92**	**1.18**

From the tensile test result, tensile strength of the specimen is increasing for pure LDPE, undrawn LDPE/r-PET and drawn LDPE/r-PET. Tensile strength of drawn LDPE/r-PET compared than pure LDPE increased by 10% while tensile

strength of drawn LDPE/r-PET compare than undrawn LDPE/r-PET increased by 6%.

For pure LDPE, undrawn LDPE/r-PET/compatibilizer and drawn LDPE/r-PET/compatibilizer, there are improvements in tensile strength. Tensile strength of drawn LDPE/r-PET/compatibilizer compared than pure LDPE increased by 18% while tensile strength of drawn LDPE/r-PET/compatibilizer compare than undrawn LDPE/r-PET/compatibilizer increased by 12%. Data for tensile strength are summarized in Figure 4.2.

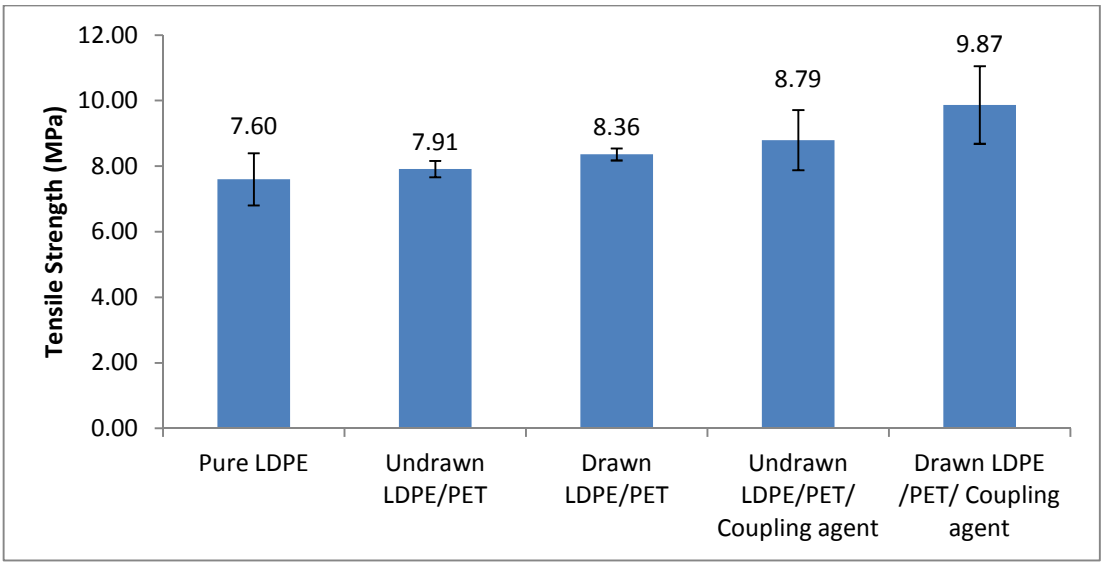

Figure 4.1: Graph of tensile strength for the tested specimens.

Morphological characteristics of the samples were studied using Scanning Electron Microscope (SEM) machine.

From the SEM micrograph, differences can be seen in the specimens. Undrawn LDPE/r-PET specimen (a) shows sphere shape of PET while drawn LDPE/r-PET specimen (b) shows fibril shape of PET. So it is proven that microfibrils are formed during the drawing process and act as reinforcement to help improving tensile strength of the blends.

To see the effect of compatibilizer towards morphology of the blend, Figure 4.2(a) and 4.2(c) is compared. With compatibilizer, it can be seen that LDPE and r-PET molecules are well blended compare than without compatibilizer. It shows that

better blending between LDPE and r-PET with aid of compatibilizer has enhanced mechanical properties of the blend by improving interfacial adhesion between the matrix and reinforcement.

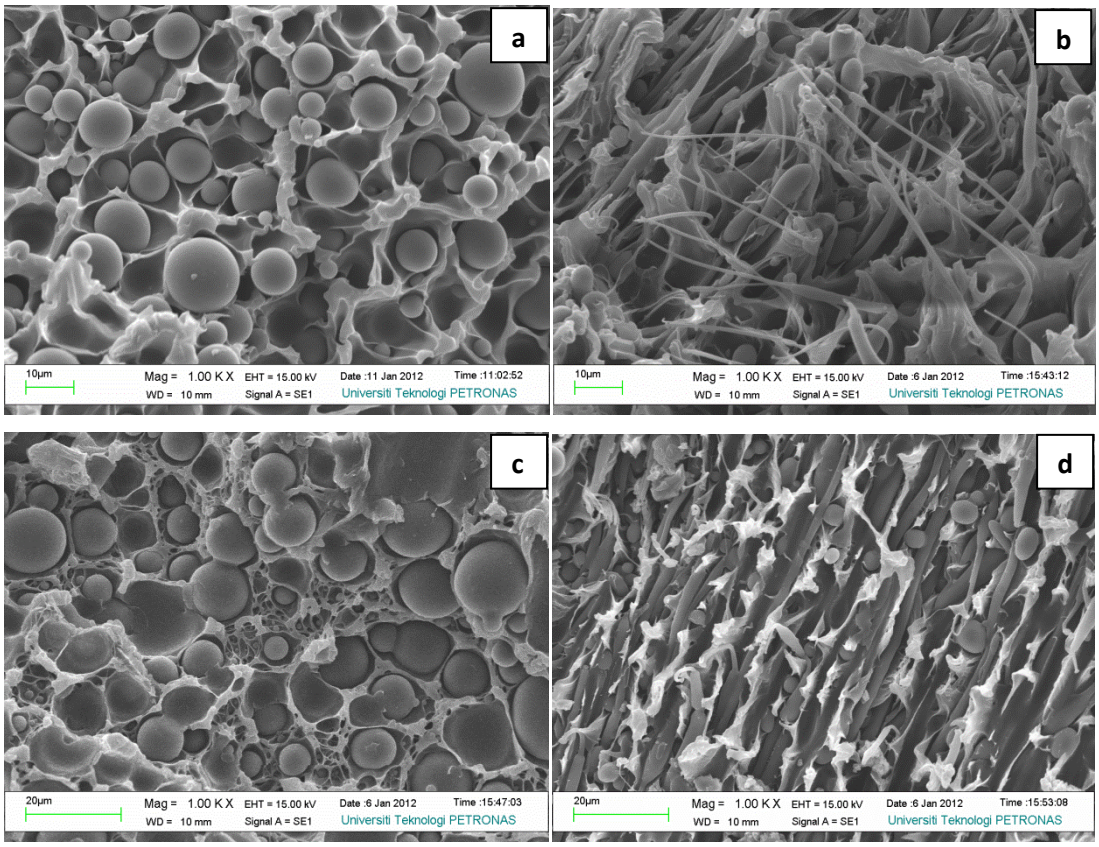

Figure 4.2: SEM pictures: (a) undrawn LDPE/r-PET (b) drawn LDPE/r-PET (c) undrawn LDPE/r-PET/coupling agent (d) drawn LDPE/r-PET/coupling agent.

4.2 Flexural Strength

Flexural test was done using three-point bending method to determine toughness of the specimens. Universal Testing Machine (UTM) is used to do this experiment. Flexural strength of the tested specimens is shown in Table 4.2.

In Undrawn LDPE/r-PET specimen, there is one sample that gives lower flexural strength compared than the other samples which is 2.58 MPa. In calculating average and standard deviation, the value is neglected as it will result in spike in the data.

The result shows improvements in flexural strengths of the specimens. Flexural strength of drawn LDPE/r-PET has improved by 13% compare than undrawn LDPE/r-PET and improved by 126% compare than pure LDPE.

Meanwhile, Undrawn LDPE/r-PET/Coupling agent sample shows 21% improvement in flexural strength compare than undrawn LDPE/r-PET and 155% improvement compare than pure LDPE. Figure 4.3 summarizes the flexural strength result.

Table 4.2: Flexural strength of the specimens.

Sample	**Flexural Strength (MPa)**				
	Pure LDPE	Undrawn LDPE/r-PET	Drawn LDPE/r-PET	Undrawn LDPE/r-PET/ Coupling agent	Drawn LDPE/r-PET/ Coupling agent
1	5.04	9.52	11.07	11.50	12.16
2	4.61	9.67	10.00	11.00	12.85
3	4.38	10.09	11.67	11.97	13.10
4	5.55	2.58	11.13	12.69	12.52
5	4.89	9.86	11.32	11.90	11.70
Avg	**4.89**	**9.78**	**11.04**	**11.81**	**12.47**
Std	**0.45**	**0.25**	**0.63**	**0.62**	**0.55**

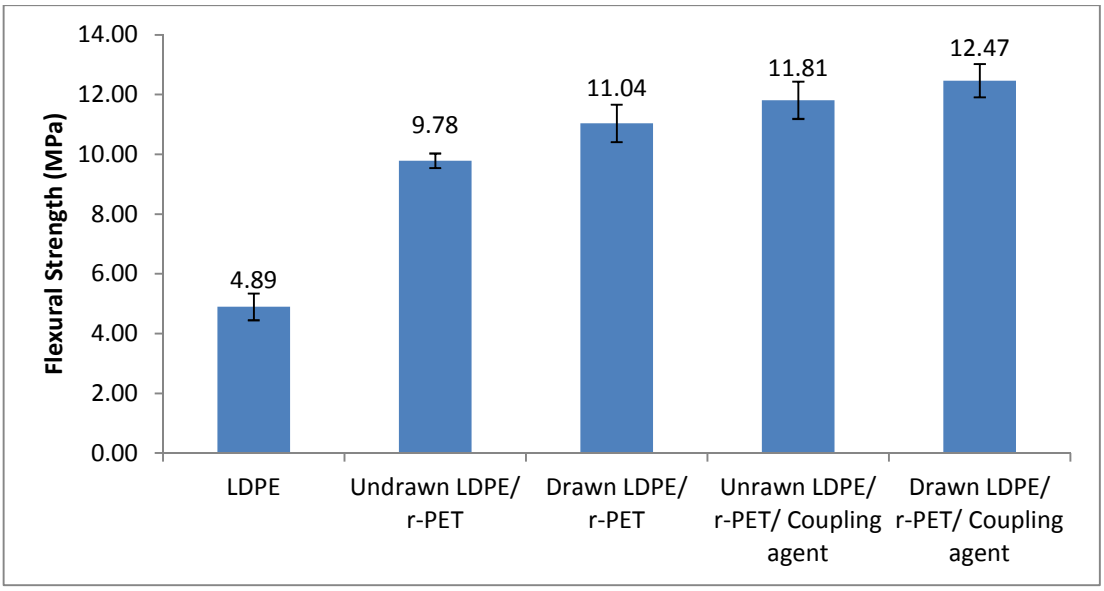

Figure 4.3: Graph of flexural strength for the tested specimens.

4.3 Impact Strength

Impact test was conducted using the Izod impact tester. Results of the test are shown in Table 4.3.

Table 4.3: Impact strength of the specimens.

Sample	Impact Strength (kJ/m^2)				
	Pure LDPE	Undrawn LDPE/r-PET	Drawn LDPE/r-PET	Undrawn LDPE/r-PET/ Coupling agent	Drawn LDPE/r-PET/ Coupling agent
1	54.18	2.37	2.20	2.13	4.09
2	50.49	2.09	2.73	2.62	3.43
3	44.02	2.29	2.46	2.48	3.20
4	64.12	2.20	2.57	2.27	3.51
5	55.50	2.16	2.40	2.53	3.14
Avg	**53.66**	**2.22**	**2.47**	**2.41**	**3.47**
Std	**7.35**	**0.11**	**0.20**	**0.20**	**0.38**

Impact strength of the Pure PE is way higher than the other specimens with r-PET. The inclusion of r-PET increased the brittleness of the sample and results in reduction of impact strength. However, comparing impact strength of undrawn LDPE/r-PET and drawn LDPE/r-PET, there is increment of 10% for the impact strength of undrawn LDPE/r-PET to drawn LDPE/r-PET. It shows the effect of microfibrillar towards toughness of the specimens.

To study the effect of coupling agent, undrawn LDPE/r-PET and undrawn LDPE/r-PET/coupling agent is compared. There is 8% of increment in impact strength between undrawn LDPE/r-PET and undrawn LDPE/r-PET/coupling agent. It shows that compatibilizer helps improving impact strength of the composite by giving better bonding between the microfibrils and the matrix. Figure 4.4 summarizes the impact strength of the specimens.

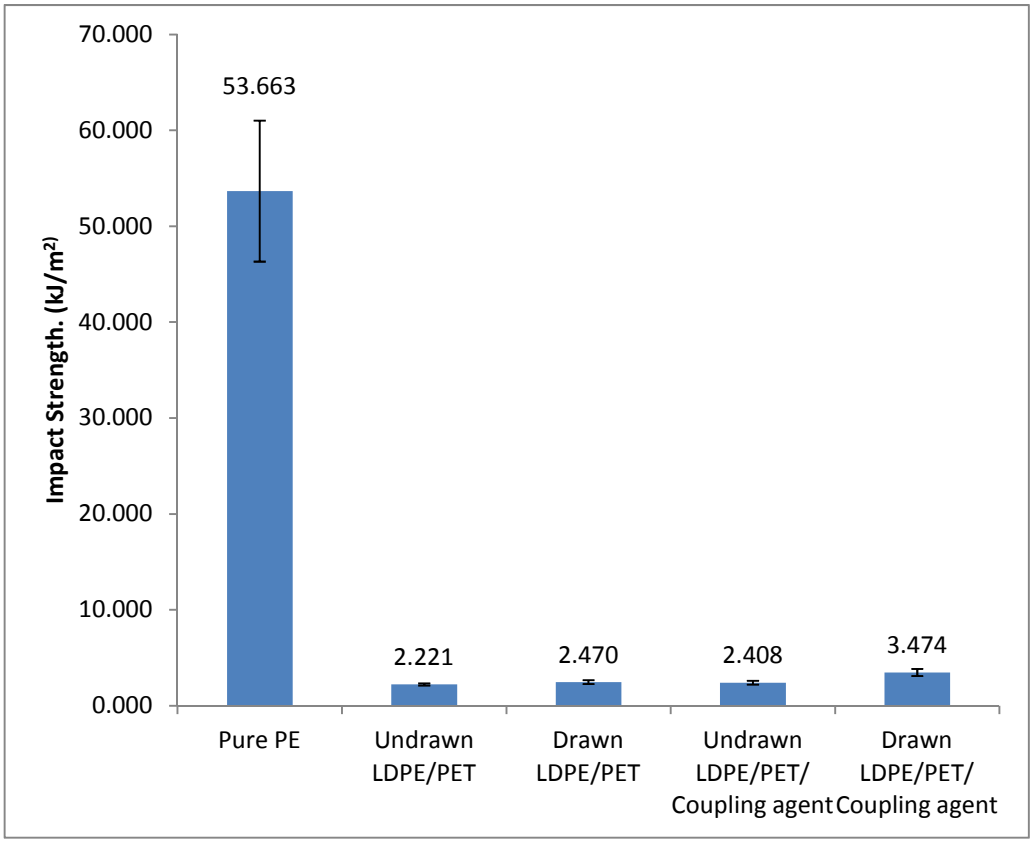

Figure 4.4: Graph of impact strength for the tested specimens.

CHAPTER 5

CONCLUSION AND RECOMMENDATION

5.1 Conclusion

Conclusions that can be drawn from this study are:

1. Microfibrillar composite has been successfully fabricated using 70/30 ratio of LDPE/r-PET.
2. Microfibrillar is proven to improve tensile strength of drawn LDPE/r-PET by 6% when compared with undrawn LDPE/r-PET and improve flexural strength by 13%. When comparing with pure LDPE, tensile strength of drawn LDPE/r-PET increased by 10% and flexural strength increased by 126%.
3. Coupling agent managed to improve tensile strength of the undrawn LDPE/r-PET/Coupling agent composite by 12% compared than undrawn LDPE/r-PET and improve flexural strength by 21%. Compared to pure LDPE, tensile strength of drawn LDPE/r-PET increased by 18% and flexural strength increased by 155%.
4. Impact strength of drawn LDPE/r-PET improved by 10% compared than undrawn LDPE/r-PET but it is reduced by 95% when compared to pure LDPE. For undrawn LDPE/r-PET/coupling agent specimen, impact strength improved by 9% compared to undrawn LDPE/r-PET but when compared to pure LDPE, impact strength value is decreased by 96%.
5. LDPE/r-PET MFCs are more suitable in applications that require tensile and flexural strengths and not suitable in applications that need high impact strength.

5.2 Recommendation

The most crucial part in this study is fabricating the microfibrillar by drawing. As UTP do not have any drawing machine, drawing is done manually and it is very challenging to produce a good drawn composite. As the result, the microfibrils are not drawn properly and some of them are in ‘cigar’ shape instead of microfibrils as seen through SEM.

In terms of mixing, the mixture of LDPE/r-PET should be extruded at least two times to produce better mixing. So that, good result can be achieved as LDPE/r-PET is well blended and well distributed in the specimen.

REFERENCES

[1] Crawford R. 1987, *Plastics Engineering*, Butterworth Heinemann

[2] Yuan Y.C., 2008, "Self-healing in Polymers and Polymer Composites. Concepts, Realization and Outlook: A review," *eXPRESS Polymer Letters* **2 (4)**: 238-250

[3] Shields, R.J., Bhattacharyya and D., Fakirov, S., 2008, "Oxygen permeability analysis of microfibril reinforced composites from PE/PET blends," *Composite* **39:** 940-949

[4] Jayanaranan, K., Thomas, S. and Joseph, K., 2008, "Morphology, static and dynamic mechanical properties of in situ microfibrillar composites based on polypropylene/poly(ethylene terephthalate) blends," *Composites* **39:** 164-175

[5] Utracki, L.A. 2002, *Polymer Blends Handbook*, Kluwer Academic Publishers

[6] Jayanarayanan, K., Thomas, S. and Joseph, K., 2011, "In situ microfibrillar blends and composites of polypropylene and poly(ethylene terephthalate): Morphology and thermal properties," *Journal of Polymer Research* **18:** 1-11

[7] Jankausaite, V., Macijauskas, G. and Lygaitis, R., 2008, "Polyethylene Terephthalate Waste Recycling and Application possibilities: a Review," *Material Science (Medžiagotyr)* **14(2):** 119-127

[8] Fakirov, S., Bhattacharyya, D. and Shields, R., 2008, "Nanofibril reinforced composites from polymer blends," *Colloids and Surfaces A: Physiochemical Engineering Aspects* **313-314:** 2-8

[9] Lei, Y., Wu, Q., Clemons, C. and Guo, W., 2009, "Phase structure and Properties of Poly(ethylene terephthalate)/High-Density Polyethylene Based on Recycled Materials," *Journal of Applied Polymer Science* **113:** 1710-1719

[10] Lei, Y., Wu, Q. and Zhang, Q., 2009, "Morphology and properties of microfibrillar composites based on recycled poly(ethylene terephthalate) and high density polyethylene," *Composites* **40:** 904-912

[11] Standard Test Method, 2004, *Tensile Properties of Plastics*, D638-03, 50-63, ASTM International.

[12] Standard Test Method, 2004, *Flexural Properties of Unreinforced and Reinforced Plastics and Electrical Insulating Materials*, D790-03, ASTM International.

[13] Standard Test Method, 2005, *Determining the Izod Pendulum Impact Resistance of Plastics*, D256-05, ASTM International.

[14] Fuchs, C., Bhattacharrya, D. and Fakirov S., 2006, "Microfibril reinforced polymer-polymer composites: Application of Tsai-Hill equation to PP/PET composites," *Composites Science and Technology* **66:**, 3161-3171

[15] Sarkissova, M., Harrats, C., Groeninckx, G. and Thomas S., 2004, "Design and charecterisation of Microfibrillar reinforced composite materials based on PET/PA12 blends," *Composites* **35:** 489-499

[16] Yi, X., Xu, L., Wang, Y., Zhong, G., Ji, X. and Li, Z., 2010, "Morphology and properties of isostatic polypropylene/poly(ethylene terephthalate) in situ microfibrillar reinforced blends: Influence of viscosity ratio," *European Polymer Journal* **46:** 719-730

[17] Li, Z., Li, L., Shen, K., Yang, W., Huang, R. and Yang M, 2003, "Transcrystalline Morphology of an in situ Microfibrillar Poly(ethylene terephthalate)/Poly(propylene) Blend Fabricated through a Slit Extrusion Hot Stretching-Quenching Process," *Macromolecular Rapid Communications* **25:** 553-558

[18] Serhatkulu, T., Erman, B., Bahar, I. and Fakirov, S., 1995, "Dynamic mechanical study of amorphous phases in poly(ethylene terephthalate)/nylon-6 blends," *Polymer* **38 (12):** 2371-2377

[19] Shields, R.J., Bhattacharrya, D., and Fakirov S., 2008, "Fibrillar polymer-polymer composites: morphology, properties and applications," *Journal of Material Science* **43:** 6758-6770

[20] Li, Z., Yang, M., Lu, A., Feng J. and Huang R., 2002, "Tensile properties of poly(ethylene terephthalate) and polyethylene in-situ microfiber reinforced composite formed via slit die extrusion and hot-stretching," *Materials Letters* **56:** 756-762

[21] Friedrich, K., Evstatiev, M., Fakirov, S., Evstatiev, O., Ishii, M. and Harrass, M., 2005, "Microfibrillar reinforced composites from PET/PP blends: processing, morphology and mechanical properties," *Composites Science and Technology* **65:** 107-116

[22] Taepaiboon, P., Junkasem, J., Dangtungee, R., Amornsakchai, T. and Supaphol P., 2006, "*In Situ* Microfibrillar-Reinforced Composites of Isostatic Polypropylene/Recycled Poly (ethylene terephthalate) System and

Effects of Compatibilizer," *Journal of Applied Polymer Science* **102:** 1173-1181

[23] Jayanaranan, K., Jose, T., Thomas, S. and Joseph, K., 2009, "Effects of draw ratio on the microstructure, thermal, tensile and dynamic rheological properties of insitu microfibrillar composites," *European Polymer Journal* **45:** 1738-1747

[24] Jayanaranan, K., Thomas, S. and Joseph, K., 2010, "Influence of cold drawing on the properties of microfibrillar structured insitu composites from PP and PET," *International Conference on Advances in Polymer Technology*, 253-256.

[25] Evstatiev, M. and Nicolov, N., 1996, "Morphology of microfibrillar reinforced composites PET/PA 6 blend," *Polymer* **37(20):** 4455-4463

[26] Zhong, G., Li, Z., Li, L. and Shen, K., 2008, "Crystallization of oriented isostatic poplypropylene (iPP) in the presence of in situ poly(ethylene terephthalate) (PET) microfibrils," *Polymer* **49:** 4271-4278

[27] Xu, X., Li, Z., Yang, M., Jiang, S. and Huang, R., 2005, "The role of the surface microstructure of the microfibrils in an electrically conductive microfibrillar carbon black/poly(ethylene terephthalate)/polyethylene composite," *Carbon* **43:** 1479-1487

[28] Fakirov, S. and Evstatiev, M., 1993, *The Interfacial Interactions in Polymeric Composites* (Editted by Akovali G.), Kluwer Academic Publisher

[29] Shields R.J., 2008, *Characterization of the mechanical and oxygen barrier properties of microfibril reinforced composites*, Ph.D Thesis, Auckland University, New Zealand

[30] Ballauri B., Trabuio M andLa Mantia F.P. *Compatibilization of Recycled Polyethyleneterephthalate/Polypropylene Blends Using a Functionalized Rubber*

[31] Vasile C. & Pascu M. 2005, *Practical Guide to Polyethylene*, Rapra Technology Limited

[32] White J. & Choi D. 2005, *Polyolefins Processing, Structure Development and Properties*, Hanser Publishers

[33] Callister W. 2007, *Material Science and Engineering: An Introduction*, John Wiley & Sons

[34] Utracki L. A. 1998, *Commercial Polymer Blends*, Chapman & Hall.

[35] Low-density polyethylene: branched form of polyethylene. [Art]. In *Encyclopædia Britannica*. Retrieved on 4 Dec 2011 from <http://www.britannica.com/EBchecked/media/2948/The-branched-form-of-polyethylene-known-as-low-density-polyethylene>.

[36] Peacock A. 2000. *Handbook of Polyethylene: Structures, Properties and Applications*. Marcel Dekker, Inc.

[37] Evstatiev, M., Fakirov, S., Krasteva, B., Friedrich, K., Covas, J.A. and Cunha, A.M., 2002, "Recycling of poly(ethylene terephthalate) as polymer-polymer composites," *Polymer Engineering and Science* **4:** 826-835

APPENDIX A

Titan Chemicals TITANLENE® LDI300YY Low density polyethylene resin

Categories: Polymer; Thermoplastic; Polyethylene; LDPE; Low Density Polyethylene (LDPE), Molded

Material Notes: Low density polyethylene for injection molding

Applications:

- Medium sized moldings
- Cosmetic containers
- Bottle closures
- Food containers

Additives: Barefoot

Advantages:

- High stiffness
- Good toughness
- Excellent gloss

Information provided by Titan Chemicals

Vendors: No vendors are listed for this material. Please click here if you are a supplier and would like information on how to add your listing to this material.

Physical Properties	Metric	English	Comments
Base Resin Density	0.920 g/cc	0.0332 lb/in³	ASTM D1505
Base Resin Melt Index	20.0 g/10 min	20.0 g/10 min	ASTM D1238

Mechanical Properties	Metric	English	Comments
Tensile Strength at Break	9.81 MPa	1420 psi	ASTM D638
Tensile Strength, Yield	11.8 MPa	1710 psi	ASTM D638
Elongation at Break	120 %	120 %	ASTM D638
1% Secant Modulus	186.32 MPa	27024 psi	ASTM D638

Thermal Properties	Metric	English	Comments
Vicat Softening Point	87.0 °C	189 °F	resin; ASTM D1525
Brittleness Temperature	-35.0 °C	-31.0 °F	ASTM D746

Processing Properties	Metric	English	Comments
Melt Temperature	160 - 240 °C	320 - 464 °F	
Mold Temperature	15.0 - 50.0 °C	59.0 - 122 °F	

APPENDIX B

DuPont Packaging & Industrial Polymers

DuPont™ Fusabond® P 613

Fusabond® resins Product Data Sheet

Description

Product Description	DuPont™ Fusabond® P613 is a anhydride modified polypropylene.

Restrictions

Material Status	• Commercial: Active
Availability	• Globally

Typical Characteristics

Uses	• Polymer Modifier
Applications	Coupling agent, short glass fiber filled PP

Typical Properties

Physical	Nominal Values	Test Method (s)	
Density ()	0.903 g/cm³	ASTM D792	ISO 1183
Melt Flow Rate (190°C/1.0kg, measured value)	49 g/10 min	ASTM D1238	ISO 1133
Melt Index (190°C/2.16kg, estimated value)	120 g/10 min	ASTM D1238	ISO 1133

Thermal	Nominal Values	Test Method (s)	
Melting Point (DSC)	162°C (324°F)	ASTM D3418	ISO 3146

Processing Information

General

Maximum Processing Temperature	300°C (572°F)

FDA Status Information	FUSABOND P613 resin conforms with the Code of Federal Regulations, Title 21, Paragraph 175.105, covering the use of adhesive interlayers in composite packages for food use. This regulation describes adhesives which may be safely used as components of articles intended for use in packaging, transporting or holding food. This regulation requires that either (1) the adhesive is separated from the food by a functional barrier, or (2) the quantity of adhesive which contacts fatty or aqueous foods does not exceed the trace amounts at the seams or edges. Customers should satisfy themselves that the food contact material is serving as a functional barrier to the adhesive.
Regulatory Information	For information on regulatory compliance outside of the U.S., consult your local DuPont representative.
Safety & Handling	As with any hot material, care should be taken to protect the hands and other exposed parts of the body when working with molten polymer.

At temperatures above300°C (572°F), these resins can evolve low concentrations of fumes. When resins are overheated, more extensive decomposition may occur. Because fumes produced during exposure to high temperatures may be combustible, exposure of overheated resin to atmospheric oxygen should be avoided if possible. Adequate local ventilation should be provided to remove the fumes from the work area.

Disposal of scrap material presents no special problems, and may be accomplished by landfill or by incineration by a properly operated incinerator. Disposal should comply with local, state, and federal regulations. Resin pellets can be a slipping hazard. Loose pellets should be swept up promptly to prevent falls.

For more detailed information on the safe handling and disposal of these resins, a Product Safety Bulletin and OSHA Material Safety Data Sheets can be obtained from the Regional Office serving you.

Read and Understand the Material Safety Data Sheet (MSDS) before using this product

Regional Centres

DuPont operates in more than 70 countries. For help finding a local representative, please contact one of the following regional customer contact centers:

Americas

DuPont Company, BMP26-2215
Lancaster Pike & Route 141
Wilmington, DE 19805 U.S.A.
Telephone +1 302-774-1161
Toll-free (USA) 800-628-6208, ext. 6
Fax +1 302-355-4056

DuPont do Brasil, S.A.
Alameda Itapecuru, 506
06454-080 Barueri, SP Brasil
Telephone +55 11 4166 8000
Fax +55 11 4166 8736

Asia Pacific

DuPont China Holding Co., Ltd.
Shanghai Branch
399 Keyuan Road, Bldg. 11
Zhangjiang Hi-Tech Park
Pudong New District, Shanghai
P.R. China (Postcode: 201203)
Telephone +86 21 3862 2888
Fax +86-21-3862-2889

Europe / Middle East / Africa

DuPont de Nemours Int'1. S.A.
2,Chemin du Pavillon Box 50
CH-1218 Le Grand Saconnex
Geneva, Switzerland
Telephone +41 22 717 51 11
Fax +41 22 717 55 00

http://fusabond.dupont.com

This data sheet is effective as of 01/11/2010 1:31PM and supersedes all previous versions.