CHARACTERIZATION AND TREATMENTS OF PINEAPPLE LEAF FIBRE THERMOPLASTIC COMPOSITE FOR CONSTRUCTION APPLICATION

(PENCIRIAN DAN PERAWATAN GENTIAN DAUN NENAS TERMOPLASTIK KOMPOSIT BAGI PENGGUNAAN PEMBINAAN)

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Universiti Teknologi Malaysia

2007

ACKNOWLEDGEMENT

Sincere thanks are indebted to Miss Suriani A.Samat as a Research assistant and to all staff and colleagues in the Department of Polymer Engineering, UTM, who have helped in any way towards the completion of this work.

We wish to express our deepest appreciation to Tuan Hj. Miskam Osman, Estate Manager, Pineapple Cannery of Malaysia Sdn Bhd., Pekan Nanas, Johor for providing pineapple leaf.

Finally, we would like to acknowledge Research Management Centre (RMC), UTM for the grant awarded.

ABSTRACT

Characterization and Treatments of Pineapple Leaf Fibre Thermoplastic Composite for Construction Application

(Keywords: Pineapple leaf fibre; characterization; treatments; polypropylene; composites; construction)

In recent years natural fibres appear to be the outstanding materials which come as the viable and abundant substitute for the expensive and nonrenewable synthetic fibre. Natural fibres like sisal, banana, jute, oil plam, kenaf and coir has been used as reinforcement in thermoplastic composite for applications in consumer goods, furniture, low cost housing and civil structures. Pineapple leaf fibre (PALF) is one of them that have also good potential as reinforcement in thermoplastic composite. It is the objective of the current research to characterize PALF and to investigate the effect of fibre treatment on the mechanical properties of PALF reinforced polypropylene (PP) composite. PALF was prepared from raw pineapple leaf. It was then chemically treated to hinder the water content. Both PP and PALF were compounded using two-roll mill machine prior to compression moulding via hot press machine to form a sheet. After forming the composite sheet, samples were prepared for tensile test (ASTM D638), flexural test (ASTM D790) and impact test (ASTM D256). Scanning Electron Microscope (SEM) was used to investigate the miscibility between the fibre and matrix. It was found that PALF contain 87.56% holocellulose, 78.11% alpha cellulose, 9.45% hemicellulose and 4.78 % lignin. The chemical constituents obtained were in the range to data reported in literatures. It was also observed that the flexural modulus and strength of treated PALF reinforced PP composite increased linearly with increment of fibre loadings. This trend was similar for impact strength where it exhibited a slight reduction at the initial stage but increased later as the fibre loading increased. The study has demonstrated that the optimum fibre loading for the best performance of the composite achieved was 30 wt%. This was clarified further by SEM where fibres and matrix have shown better miscibility at 30 wt% of treated PALF.

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ABSTRAK

Pencirian dan Perawatan Gentian Daun Nenas Termoplastik Komposit bagi Penggunaan Pembinaan

(Katakunci: Gentian Daun Nenas; Pencirian; Perawatas; polipropilena; komposit; pembinaan)

Sejak tahun kebelakangan ini, gentian asli muncul menjadi bahan sisa yang dapat menggantikan gentian sintetik yang mahal harganya. Gentian asli seperti sisal, gentian daripada pisang, jut, kelapa sawit, kenaf dan sabut kelapa telah digunakan sebagai bahan pentetulang komposit termoplastik bagi penggunaan seperti perabot, rumah kos rendah dan struktur binaan. Gentian Daun Nenas (PALF) adalah salah satu daripada gentian yang juga mempunyai potensi untuk dijadikan sebagai bahan pentetulang dalam termoplastik komposit. Adalah menjadi tujuan penyelidikan ini untuk membuat pencirian keatas daun nenas dan mengkaji kesan rawatan ke atas sifat-sifat mekanikal PALF bertetulang polipropilena komposit. Gentian telah disediakan daripada daun nenas yang diambil daripada ladang. Gentian kemudiannya telah dirawat sebelum diproses. Kedua-duanya iaitu polipropilena (PP) dan PALF telah disebatikan dengan menggunakan mesin penggiling berkembar sebelum dimasukkan ke dalam mesin acuan mampatan untuk membentuk kepingan. Selepas pembentukan kepingan komposit, sampel telah disediakan untuk ujian regangan (ASTM D638), ujian lenturan (ASTM D790) dan ujian mampatan (ASTM D256). Mikroskop Imbasan Elektron (SEM) telah digunakan untuk menyelidik kebolehcampuran antara gentian daun nenas dan matriks polipropilena. Adalah didapati bahwa PALF mengandungi 87.56% holocellulose, 78.11% alpha cellulose, 9.45% hemicellulose dan 4.78 % lignin. Kandungan kimia yang didapati adalah dalam julat seperti yang dilaporkan dalam literatur. Diperhatikan juga modulus lenturan dan kekuatan rawatan daun nenas bertetulang komposit PP bertambah secara lurus dengan pertambahan gentian. Keputusan ini adalah sama dengan kekuatan hentaman di mana pada peringkat awalnya ia menunjukkan penurunan kekuatan tetapi meningkat setelah komposisi gentian bertambah. Kajian ini telah menunjukkan bahwa komposisi gentian yang optima untuk menghasilkan bahan komposit adalah pada 30 % pecahan berat. Ini telah dibuktikan dengan analisa SEM di mana gentian dan matriks telah menunujukkan kebolehcampuran yang baik pada kadar 30 peratus berat PALF yang dirawat.

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CHAPTER 1

INTRODUCTION

1.1 Introduction

Agriculture is an important sector in Malaysian economy. Traditionally, agricultural materials have been shipped away for processing, or disposed of post-harvest. Diversification of the industry is crucial in encouraging economic stability and growth. Value-added processing would helps in agricultural diversification. Pineapple Leaf Fibre (PALF), the subject of the present study, is a waste product of pineapple cultivation. Hence, pineapple fibre can be obtained for industrial purposes without any additional cost.

Traditional plastic materials are reinforced by glass fibers, which are both expensive and harmful to the environment. A fibre based biocomposite material contains polymers reinforced with natural fiber. There are a number of advantages of using natural fibers in biocomposites, among which are: a) natural fiber will make the material partially biodegradable; b) glass fiber is relatively expensive to make; c) natural fibre, in this research is PALF is currently disposed of by burning; and d) PALF is a low cost, low density and low energy consumption. Over the past decade, cellulosic fillers have been of greater interest as they give composites improved mechanical properties compared to those containing non-fibrous fillers. In recent years, thermoplastic materials have been increasingly used for various applications (Folkes, 1982).

Natural fiber-reinforced thermoplastic composites form a new class of materials which seem to have good potential in the future as a substitute for wood-based material in many applications. However, lack of good interfacial adhesion and poor resistance to
moisture absorption makes the use of natural fiber-reinforced composites less attractive. Various fiber surface treatments like mercerization, isocyanate treatment, acrylation, latex coating, permanagante treatment, acetylation, silane treatment and peroxide treatment have been carried out which may result in improving composite properties. Research on a cost effective modification of natural fibers is necessary since the main attraction for today's market of biocomposites is the competitive cost of natural fiber. Interfaces play an important role in the physical and mechanical properties of composites. Reinforcing fibers are normally given surface treatments to improve their compatibility with the polymer matrix. This research attempts to address the following question: do chemical treatments of PALF have any influence on the composite properties?

The creation of fiber-reinforced composites is a multi-step process. First the PALF is extracted from its leaf and then, chemical treatment is used to reduce it to its fibrous form. Next it is chopped to appropriate size and combined with synthetic polymer materials. A series of steps including extrusion and plastic molding techniques
are used to develop the final product. PALF reinforced thermoplastic composite has material properties similar to that of conventional plastic products. The environmental benefits of the fiber-reinforced composite are appealing to producers, consumers, and industry alike. The goal of this study is to characterize and determine the effects of treatment on the performance of PALF reinforced polypropylene composites.

1.2 Objective

The specific objectives of this research were to:

i. To characterize pineapple leaf fibre

ii. To investigate the effect of surface fibre treatments on the mechanical strength of pineapple leaf fibre polypropylene composite

iii. To study the fibre matrix interaction by scanning electron microscopy (SEM)

1.3 Scopes of Research

The scopes of research were:

(i) Preparation of short pineapple leaf fibre (PALF)

(ii) Characterization of PALF

(iii) Treatment of fibre prepared

(iv) Addition of coupling agent

(v) Compounding of PALF and PP using two-roll mill machine

(vi) Hot Press

(vii) Testing to find out mechanical properties

- Tensile Test (ASTM D638)
- Flexural Test (ASTM D790)
- Impact Test (ASTM D256)

(viii) Scanning Electron Microscope (SEM) to study fibre matrix interaction

CHAPTER 2

LITERATURE REVIEW

2.1 Introduction

Modification of properties can be done by the physical combination of two polymers or by the physical combination of a polymer with a non-polymeric component. One type of the methods is through forming composite material. Composites are formed by combination of a polymer of some type non-polymeric solid or by combination of some types of engineering materials. Commonly, composites tend to have characteristics such as high strength; high modulus; low density; and excellent resistance to fatigue, creep, creep rupture, corrosion and wear. Table 2.1 lists some advantages and disadvantages of composites.

Table 2.1: Advantages and Disadvantages of Commercial Composite (Peter, 2002)

Advantages	Disadvantages
▪ Weight reduction	▪ Cost of raw materials and fabrication

▪ High strength- or stiffness- to-weight ratio ▪ Tailorable properties: can tailor strength or stiffness to be in the load direction ▪ Redundant load paths (fibre to fibre) ▪ Longer life (no corrosion), better fatigue life ▪ Lower manufacturing costs ▪ Inherent damping ▪ Increased (or decreased) thermal or electrical conductivity	▪ Transverse properties may be weak ▪ Matrix weakness, low toughness ▪ Matrix subject to environmental degradation ▪ Difficult to attach ▪ Analysis for physical properties and mechanical properties difficult, analysis for damping efficiency has not reached a consensus ▪ Non-destructive testing tedious

From Table 2.1, application of typical or commercial composites still limited due to the economic factor. Since these composites used fibre glass or other engineering material as the reinforcement, the cost of raw materials and fabrication will be high. This is very obvious in industrial field that required advance composite material, for instance aeronautic and marine engineering.

In general, polymeric composites are formed by combining fibres and polymer resin which also known as fibre reinforced plastic (FRP). The term polymer refers as long chain molecule that is composed of a large number of repeating units of identical structure. This covalent bonding is formed via polymerization process and can have shape of linear or even more complicated networking.

Polymer can be divided into two major groups based on their thermal processing behaviors: thermoset and thermoplastic. Thermoplastic refers to polymer that can be melt processed by a variety of methods, including extrusion and moulding. These include polyethylene, polypropylene, polystyrene and polyvinyl chloride. On the other hand, thermoset are polymer whose individual chains have been chemically linked by covalent bonds during polymerization or by subsequent chemical or thermal treatment. Once formed, the crosslinked networks resist heat softening, creep and solvent attack. Principle thermosets are epoxies, polyesters and formaldehyde-based resins.

Judging from the trade off between stable thermosets and recyclability of thermoplastic, the main concern of this study is on thermoplastic. Polypropylene (PP)

has been chosen as the matrix of composite as it has low processing temperature (below 230˚C) which will not degrade the fibre.

PP is one of the most successful commodity synthetic polymers. In volume terms, PP takes the fourth place after polyester, polyamide and acrylic fibres. It is widely used because of its low density (0.905 g/cm^3), high crystallinity and also high stiffness and hardness. Table 2.2 is the properties of five commercial materials by the same manufacturer which are of approximately the same isotactic content but which differ in molecular weight. Increase in melt flow index (MFI) means decrease in molecular weight.

Table 2.2: Some Mechanical and Thermal Properties of Commercial Polypropylene (Brydson, 1999)

Property	Test Method	Homopolymer		
Melt Flow Index (MFI)	(a)	3.0	0.7	0.2
Tensile Strength				
(lb in^{-2})	(b)	5000	4400	4200
(MN/m^2)		34	30	29
Elongation at Break (%)	(b)	350	115	175
Flexural Modulus				
(lb in^{-2})	-	190000	170000	160000
(MN/m^2)		1310	1170	1100
Brittleness Temperature (˚C)	I.CI. /ASTM D746	+15	0	0
Vicat Softening Point (˚C)	BS 2782	145-150	148	148
Rockwell hardness (R-scale)	-	95	90	90
Impact Strength (ft lb)		10	25	34
(J)	(c)	13.5	34	46

(a) Standard polyethylene grader: load 2.16kg at 230˚C
(b) Straining rate 18 in/min
(c) Falling weight test on 14 in diameter moulded bowls at 20˚C

From Klaus-Peter Mieck's perspective (1999), the grade of matrix forming PP has less effect on the tensile and flexural properties of the composites. Generally, PP of fineness between 1.7-6.7 dtex with lengths of 40-60 mm is considered suitable. Even PP obtained from recycling processes can also be used.

2.2 Current Trend of Composite

In this new era of technology, availability of bio-based composites offer the opportunity for environmental gains, reduced energy consumption, light weight, insulation and sound absorption properties, reduction in volatile organic emissions, and reduction in the dependence on petroleum based and forest product based materials. The development of sustainable materials as an alternative for petroleum based materials is being studied to decrease the dependence on oil or petroleum. This also can reduce carbon dioxide emissions and to generate a more economic opportunity for the agricultural sector globally. Figure 2.1 shows the outlook of application of bio-based composites in United State of America. It is forecasted that the introduction of bio-based composites will soon alter the global trend of composite and serves as a better choice when selecting the appropriate construction materials.

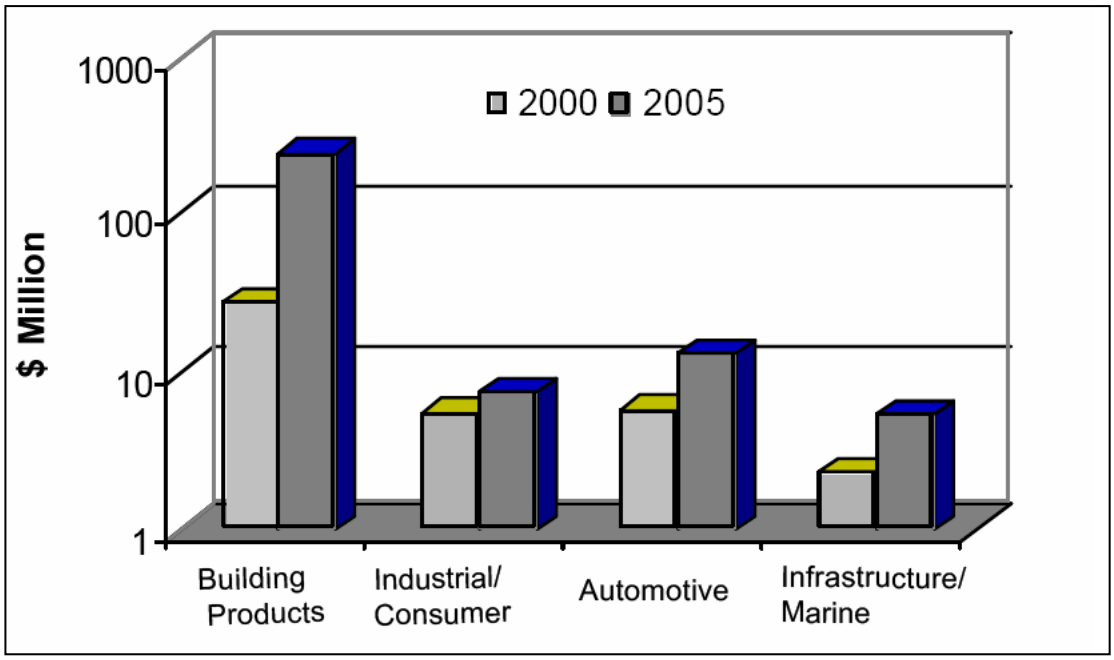


Figure 2.1: Growth Outlook for Bio-based Composites by Application in United State, 2000-2005 (Drzal *et al.*, 2003)

2.3 Natural Fibre

Bio-based composite in a more precise way refers to natural fibre reinforced composite. There is a drastic growing in the use of natural fibres as reinforcing components with petroleum and thermoplastics as well as thermosets. Advantages of these natural fibers over traditional reinforcing fibers such as glass and carbon are low cost, low density, acceptable specific properties, ease of separation, enhanced energy recovery and biodegradability. They also reduce wear on processing machinery and reduced health hazard. In the central stage of material science, natural fibres appear to be the outstanding materials which come as the viable and abundant substitute for the expensive and nonrenewable synthetic fibre. Thermoplastics have the added advantage such as recyclability however thermosets have the necessary mechanical properties for other applications. Drzall *et al.* (2003) claimed that bio-composites derived from natural fibers and petroleum-based thermoplastics or thermosets are not fully environmentally friendly because matrix resins are non-biodegradable but the bio-based content of the final composite material falls within the definition of bio-based materials. They maintain a balance between economics and environment allowing them to be considered for applications in the automotive, building, furniture and packaging industries. Markus Kaup and his colleagues (2003) have made an overall survey on natural fibre application in automotive industries. Figure 2.2 is one of their survey for various natural fibres.

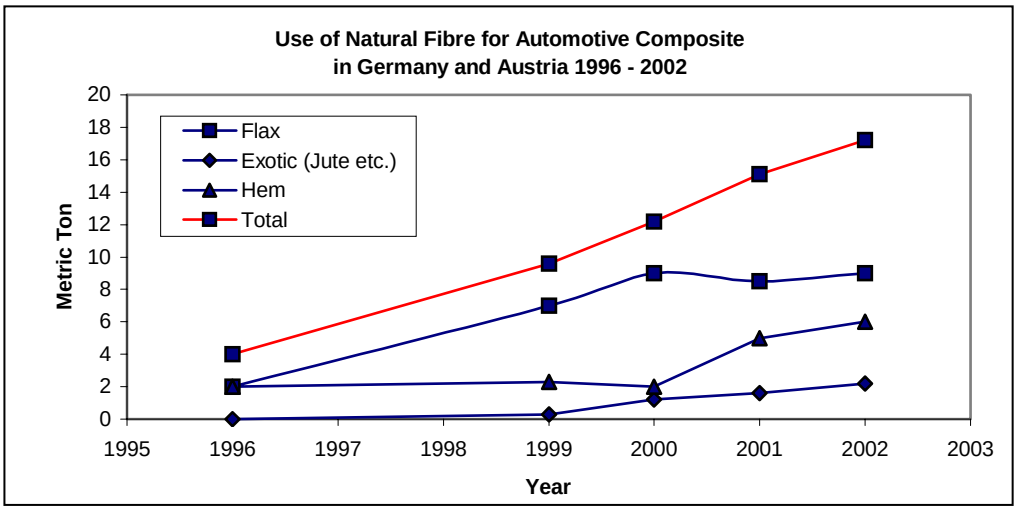


Figure 2.2: Use of Natural Fibre for Automotive Composite in Germany and Austria 1996-2002 (Markus Kaup *et al.*, 2003)

According to Figure 2.3, the cellulose based fibres can be classified to wood fibres and non-wood fibres. In non-wood fibres, they can be further classified into straw, plant (such as bast, leaf and seeds) and grass. As for the other wood fibre category, it is also quite important in the composite industries as reinforcement.

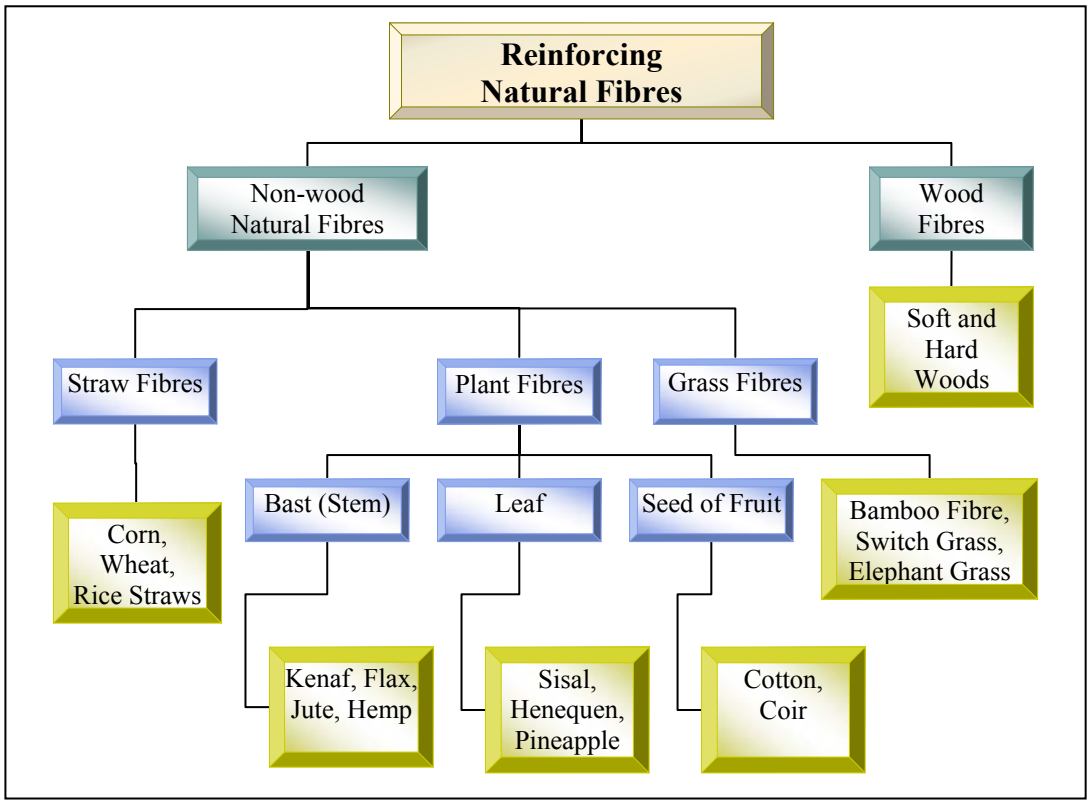


Figure 2.3: Categories of Natural Fibres

Figure 2.4 shows some types of reinforcing natural fibres that are normally been used nowadays. There are numerous researches done on natural fibres reinforced composites such as kenaf fibre, oil palm fibre, vegetable fibre, bamboo fibre, jute fibre, sisal fibre, coconut fibre and pineapple leaf fibre.

Figure 2.4: Various Types of Natural Fibres

In Figure 2.5, the tensile strength and Young's modulus of natural fibre reinforced PP-composites were studied and compared to those achieved by glass fibre (GF) reinforcement at the same reinforcement content. Since the tensile strength for these natural fibres are low, the composites reach only of about 60 % of the tensile strength of the GF-PP composites. However the Young's moduli are commonly higher than those of GF-PP composites. By considering the economy factors where the natural fibre are abundant and are relatively cheap compare to engineering glass fibre, natural fibres have their significant important in the composite field as reinforcement.

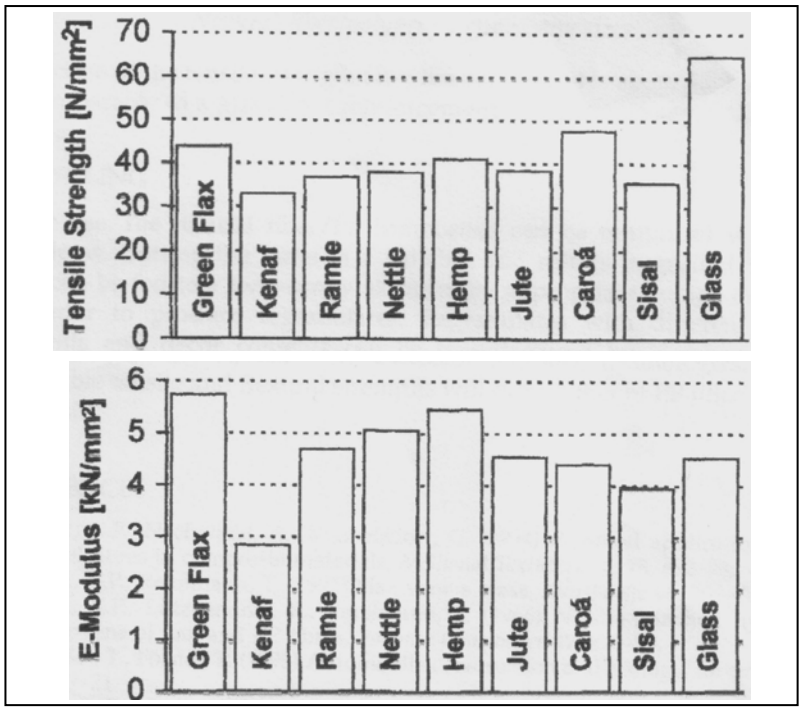


Figure 2.5: Tensile Strength and Young's Modulus of Natural Fibre and Glass Fibre Reinforced PP (Fibre Content of 30 wt %) (Klaus-Peter Mieck, 1999)

There are several factors affecting fibre properties and one of the most significant factors is the chemical properties. Rowell *et al.* (2000) has made a wide coverage on the characterization and factors affecting fibre properties. They stated that a high aspect ratio (length / width) is important in agro-based fibre (natural fibre) composites as it gives an indication of possible strength properties. A few of vast array of fibre structures that exist in the plant were shown in their work. Major differences in structure such as density and cell wall thickness, did result in differences in physical properties.

Quite a number of researches are carried out for sisal fibre reinforced polymer composites. Kuruvilla Joseph *et al.* (1999) has reviewed on work published in the field of sisal fibre reinforced polymer composites with special reference to the structure and properties of sisal fibre, processing techniques and the physical and mechanical properties of composites. The characteristic of sisal fibres depend on the properties of the individual constituents, the fibrillar structure and the lamellae matrix. In comparison with polystyrene (PS) and low density polyethylene (LDPE), PP has found to be a good matrix for sisal polyolefin composites. Since PP is more crystalline than LDPE, the increase in tensile strength by the addition of sisal fibre is less in the case of PP compared to LDPE. However the strength of the composite formed by the addition of fibre is more in the case of PP compared to LDPE especially at high fibre loading.

Herrera-Franco and Valadez-González (2004) focused on mechanical properties of continuous henequen fibres (*Agave fourcroydes*)-reinforced high density polyethylene (HDPE) composites. The fibre matrix interaction were changed with surface modification of the fibre, first to increase the area of contact and to further expose the cellulose micro fibrils and then to improve fibre wetting and impregnation. Chemical interaction was also promoted by using a silane coupling agent solution. This study has shown that the fibre surface modification had a more notorious effect on the strength properties in the perpendicular direction to the fibre where the improvements were above 50% with respect to the untreated fibre composite. The introduction of surface treatment shifted the failure mode from

interfacial failure to matrix failure. They concluded also silane has served well in enhancing adhesion between fibre and matrix. This is proven when treated fibres are still cover with layer of polymer even after failure in micro-photographs.

Rozman and his colleagues (1999) studied on the use of coconut fibre and glass as reinforcements in PP hybrid composites. The incorporation of both coconut fibre and glass fibre into the PP matrix has resulted in the reduction of flexural, tensile and impact strengths compared to unreinforced PP. This phenomenon appeared because there is more incompatibility between the fibres and the PP matrix as well as the irregularity in fibre size. As a result, the stress transfer in matrix is not efficient and lead to lowering in the strength of the material. By increasing the fibre loading, the tensile and flexural moduli have been improved. The result also indicated that more bio-fibres could be incorporated in hybrid composites, which would give the same range of properties s the composites with higher loading of glass fibres.

The study by Mubarak and Idriss Ali (1999) was based on wood-plastic composite (WPC). Low-grade wood (soft wood like kadom, simul and mango) was reinforced by monomer of methyl methacrylate (MMA) to achieve the quality of high-grade wood. The effect of additives like urea and N-vinyl pyrrolidone (NVP) was also considered. Lastly the authors concluded that there has been a significant amount of enhancement of polymer loading (PL) in the wood samples upon incorporating additives into the bulk monomer MMA. The crosslink density has increased significantly. Besides, MMA has improved the tensile strength, bending strength and compression strength of low-grade wood.

2.4 Pineapple Leaf Fibre (PALF)

Pineapple Leaf Fibre (PALF) serving as reinforcement fibre in most of the plastic matrix has shown its significant role as it is cheap, exhibiting superior properties when compared to other natural fibre as well as encouraging agriculture-

based economy. PALF is multi-cellular and lignocelluloses materials extracted from the leave of plant *Ananas cosomus* belonging to the *Bromeliaceae* family by retting (separation of fabric bundles from the cortex). PALF has a ribbon-like structure and is cemented together by lignin, pentosan-like materials, which contribute to the strength of the fibre (George *et al.*, 2000). Figure 2.6 shows that the PALF is a multicellular fibre like other vegetable fibres. Their study also found that the cells in this fibre have average diameter of about 10 μm and mean length of 4.5 mm with aspect ration of 450. The thickness of the cell wall (8.3 μm) lies between sisal (12.8 μm) and banana leaf fibre (1.2 μm). The excellent mechanical properties of PALF are associated with this high cellulose and low microfibrillar angel. Table 2.3 indicates the physical and mechanical properties of PALF obtained from South India Textile Research Association (SITRA), Coimbatore, India.

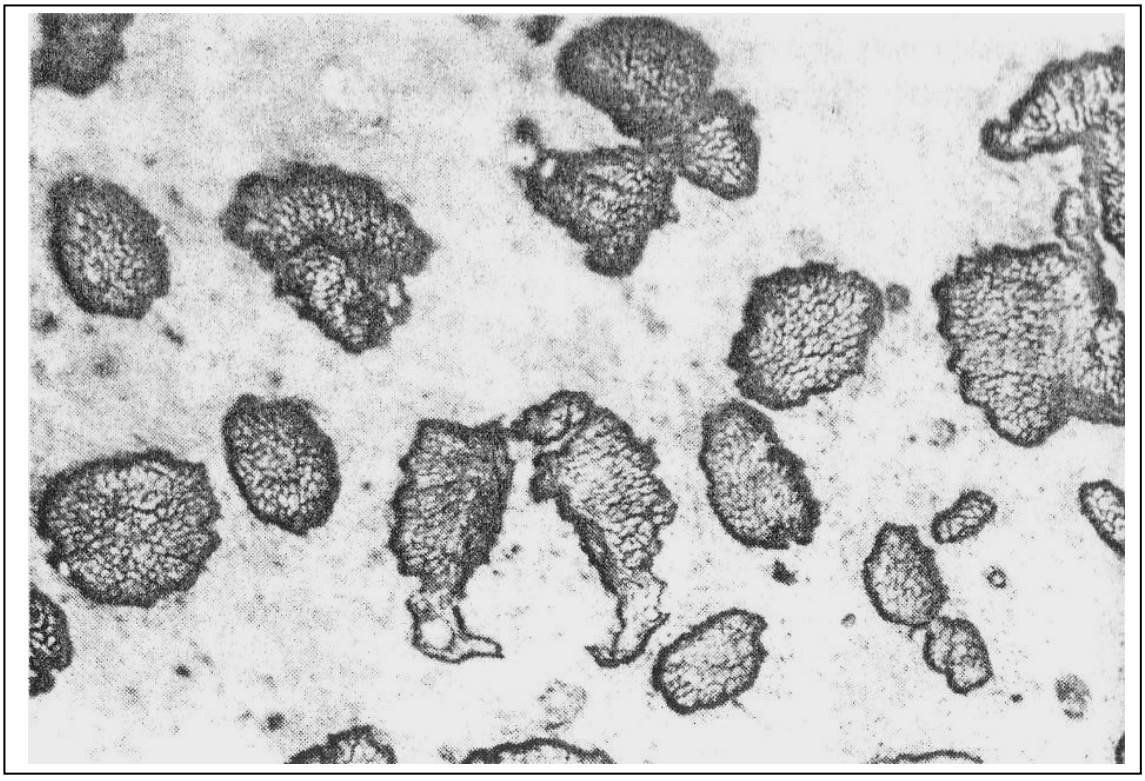


Figure 2.6: Optical Micrograph of Cross Section of PALF (× 160 magnification) (Mukherjee *et al.*, 1986)

Table 2.3: Physical and Mechanical Properties of PALF from SITRA (George *et al.*, 1995, 1998; Uma Devi *et al.*, 1997)

Properties	Value

Density	(g/cm^3)	1.526
Softening Point	(˚C)	104
Tensile Strength	(MPa)	170
Young's Modulus	(MPa)	6260
Specific Modulus	(MPa)	4070
Elongation at Break	(%)	3
Moisture regain	(%)	12

Arib and his colleagues (2004) have provided an overview of the development, mechanical properties and uses of PALF reinforced polymer composites. Both the thermosets and thermoplastic resins have been used as matrices for this natural fibre. Short PALF are mostly used in the various researchers discussed. This literature review concluded some future research that might draw PALF to a more developed area. They proposed that the major study can focus on long PALF; manufacturing such as autoclave moulding, vacuum bag moulding as well as resin transfer moulding (RTM); possible uses of PALF composites and also the study on properties can be further extended to creep, fatigue, physical and electrical properties.

There are a lot of researches done on PALF and most of them reported that incorporation of this fibre will further enhance the properties of the composite. George *et al.* (1995) studied the short PALF reinforced LDPE composites. It stressed on mechanical properties of composite prepared by two methods, namely the melt mixing and solution mixing. The influences of fibre length, fibre loading and fibre orientation have also been evaluated. Besides, the fibre breakage and damage during processing were analyzed from fibre distribution curve and optical and scanning electron micrographs. As a summary, the best optimum parameters for melt mixing are with mixing time of 6 min, rotor speed of 60 rpm and mixing temperature of 130˚C while 6mm of fibre length is the most suitable length to enhance good properties in solution mixing. In comparison of these two preparation methods, melted-mixed composites showed lower properties than solution-mixed composites due to the extensive fibre damage and breakage during melt mixing. The recyclabilty and reprocessability have also been reported. Recyclability of the

composites was found to be very good; its properties remain constant up to third extrusion. This is beyond the marginally decrease of property due to thermal effect and degradation of the fibre.

Uma Devi *et al.* (1997) investigated the mechanical behavior of PALF reinforced polyester composites as a function of fiber loading, fiber length, and fiber surface modification. Tensile strength and modulus of this thermoset composite were found to increase linearly with fiber content. The impact strength was also found to follow the same trend. However in the case of flexural strength, there was a leveling off beyond 30 wt % fiber content. A significant improvement in the mechanical properties was observed when treated fibers were used to reinforce the composite. The best improvement was observed in the study of silane A-172-treated fibre composites. Uma Devi and his members summarized that the PALF reinforced polyester composites exhibit superior mechanical properties when compared to other natural fiber polyester composites.

The study of melt rheological behaviours of short PALF-LDPE was reported by George *et al.* (1996a). The effect of fibre loading, fibre length and fibre treatment in the rheology aspect was investigated. In general, the viscosity of PALF-LDPE composite increased with fibre loading due to an increased hindrance to the flow. It was also found that the viscosity of the flow decreases with increase of temperature but not for treated fibre composite. Crosslinking happens in composite at higher temperature.

Works by George *et al.* (1996b) emphasized on the effect of fibre loading and surface modification to thermogravimetric and dynamic mechanical thermal analysis of PALF-LDPE composite. At high temperature (350˚C where cellulose decomposes), PALF degrades before the LDPE matrix. However the storage modulus *E'* increased with increase of fibre loading in dynamic mechanical thermal analysis. It was also found that improved interaction exerted by the chemical treatments makes the composition more mechanically and thermally stable than the untreated fibre composite. By the way, dynamic moduli increased with increasing frequency due to the reduced segmental mobility.

Research by George and his team (1998) shifted their interest of study on PALF-LDPE to the effects of environment. The influence of water environment on the sorption characteristics of PALF-LDPE was the main subject of their study. The effects of fiber loading, temperature and chemical treatment on the sorption behavior were evaluated. There are four chemical treatment carried out for PALF fibre which are alkali treatment (sodium hydroxide, NaOH), silane treatment (silane A172), isocyanate (poly(methylene) poly(phenyl) isocyanate, PMPPIC) and peroxide (benzoyl peroxide, BPO; dicumyl peroxide, DCP). Among these various treatments, the degree of water absorption was observed to be increased in the order PMPPIC < BPO < Silane < NaOH < DCP < Untreated. In other word, the interfacial interaction increased in the order of Untreated < DCP < NaOH < Silane < BPO < PMPPIC. Actually the fibre-matrix bonding becomes weak with increasing moisture content, resulting in interfacial failure.

Short PALF is not just limits its function as reinforcement to thermoplastic or thermoset but also to the extent of elastomer (natural rubber, NR). Timir Baran Bhattacharyya *et al.* (1986) investigated the effect of PALF on natural rubber with respect to fibre-rubber adhesion; anisotropy in physical properties; processing characteristic; ageing resistance; and comparative changes in physical properties and processing characteristics between High Abrasion Furnace (HAF) carbon black reinforced rubber. To conclude, they stated that the addition of PALF to NR increased the shore A hardness and decreased the elongation at break. Carbon black with PALF reinforced NR composite gave high hardness, low elongation, moderate tensile strength and moderate flex resistance.

In hybrid composite which consist of more then two materials, Mohanty *et al.* (2000) focused on chemical modification of PALF with graft copolymerization of acrylonitrile onto defatted PALF. The graft copolymerization of acrylonitrile (AN) onto defatted PALF was studied using combination of cuprum sulfate ($CuSO_4$) and potassium periodate (KIO_4) as an initiator in an aqueous medium. The effects of these substances' concentration, time, temperature, amount of some inorganic salts and organic solvents on the graft yield were reported. In conclusion, the writers

stated that the combination of [Cu^{2+}] ion and [IO^{4-}] ion produced optimum grafting at certain condition. Neither KIO_4 nor $CuSO_4$ alone can induce the polymerization of AN to the PALF surface. All in all, grafting improves the thermal stability of PALF.

As a whole, a numbers of researchers agreed that PALF has improved the properties of various composites. In the following sections, some major mechanical properties will be elaborated, namely tensile, flexural and impact properties.

2.5 Tensile Properties of PALF Composite

Tensile properties are some of the most widely tested properties of natural fibre reinforced composites. Recently, investigation for the PALF reinforced composite's tensile properties have covered the effect of mixing condition for melt mixing, condition for solution mixing, fibre length, fibre loading, chemical treatment, fibre orientation, water absorption and weathering effect. Noteworthy studies of tensile properties of PALF reinforced composites are Uma Devi *et al.* (1997), George *et al.* (1995) and George *et al.* (1997).

Uma Devi *et al.* (1997) observed that there were changes in tensile properties with fibre length and fibre loading of PALF in polyester composites. Table 2.4 shows that the most outstanding Young's modulus achieved for PALF reinforced polyester is at fibre length of 30 mm with value of 2290 MPa. The author believed that the decrease in the strength of the fibres above a fibre length of 30 mm can be explained as fibre entanglements occur above an optimum size of fibre. As for fibre loading, Table 2.5 compares the tensile properties with the effect of fibre loading. Basically, addition of fibre loading increased the Young's modulus. Addition of 40 wt % of fibre increased the modulus by 340 %. For tensile strength, as an overall, the properties increased when the fibre loading increased. However the addition of 10 wt % fibre showed a slight decrease in tensile strength. This is attributed by the fact that small amount of fibre acts as flaws.

Table 2.4: Effect of Fibre Length on Tensile Properties of PALF-reinforced Polyester Composites (Fibre Content of 30 wt %) (Uma Devi *et al.*, 1997)

Fibre Length (mm)	Young's Modulus (MPa)	Tensile Strength (MPa)	Elongation at Break (%)
5	815	15.6	3.0
10	1870	35.0	3.0
20	1990	39.2	3.0
30	2290	52.9	3.6
40	1970	38.4	3.0

Table 2.5: Variation of Tensile Properties of PALF-reinforced Polyester Composites as a Function of Fibre Loading (Fibre Length of 30mm) (Uma Devi *et al.*, 1997)

Fibre Content (wt %)	Young's Modulus (MPa)	Tensile Strength (MPa)	Elongation at Break (%)
0	580	20.6	1.6
10	1770	17.1	1.3
20	1830	40.0	3.0
30	2290	52.9	3.6
40	2520	63.3	5.0

George *et al.* (1995) compared the properties exhibited by melt-mixing and solution mixing methods to produce PALF-LDPE composites. From Figure 2.7(a), it can be seen that when the mixing time is less; tensile strength and Young's modulus are decreasing because of the ineffective mixing and poor dispersion of the fibre in LDPE. On the other hand, as mixing time increases, the tensile strength increases

and have the optimum mixing time of 6 minutes. The orientated composite exhibits higher tensile properties as shown compared to the randomly oriented composite. Figure 2.7(b) describes that as rotor speed is higher, the tensile properties increased. However there is a level off at the peek point of 60 rpm. The increased rotor speed to 80 rpm shows reduction in strength occurs due to the fibre breakage at higher rotor speed.

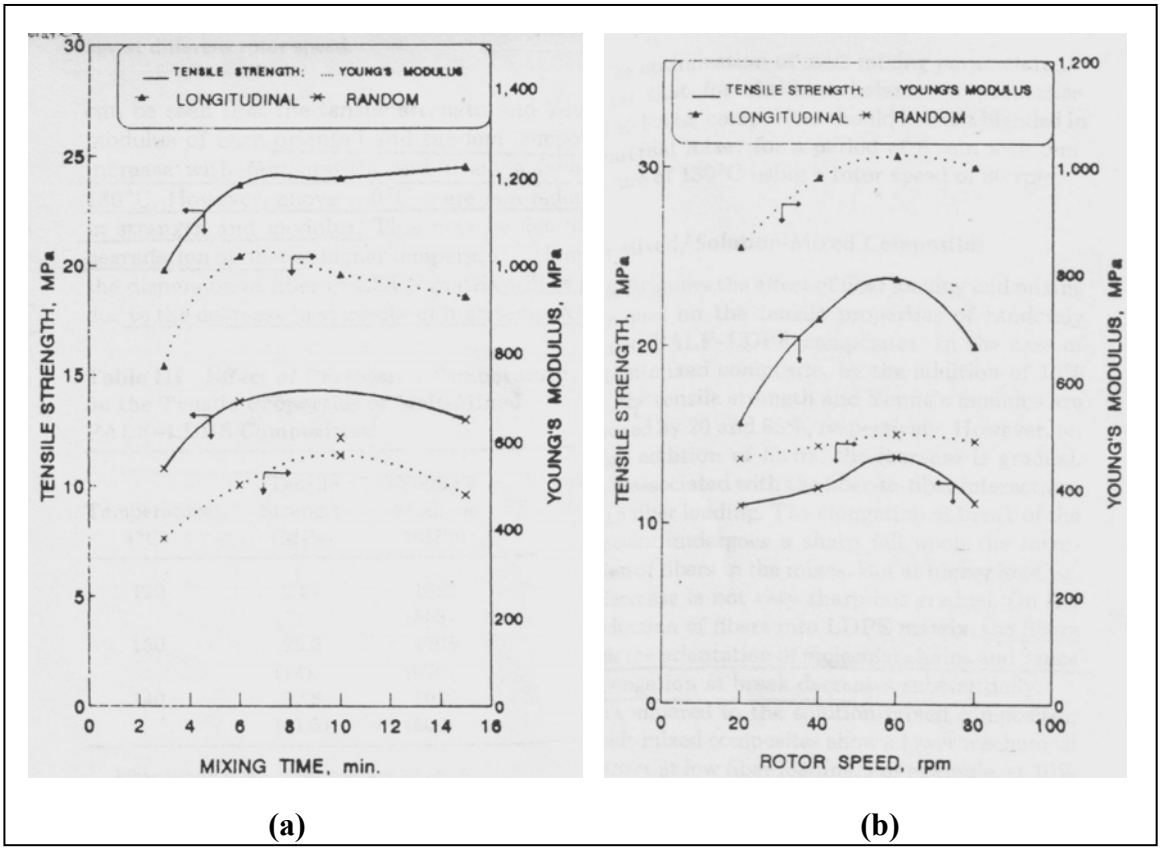


Figure 2.7: Graphs of Effect of Tensile Strength and Modulus for Melt-mixed Composites (using Brabender Plasticoder) with (a) mixing time and (b) rotor speed. Both with fibre content 30 wt %. (George *et al.*, 1995)

Table 2.6 summarizes that pineapple- and sisal-fibre- filled composites have comparable mechanical properties. In longitudinally oriented PALF-LDPE, the addition of 10 wt % fibre causes an increase of about 92 % in tensile strength for LDPE whereas in sisal composites the corresponding value cited 83 % only. However sisal-LDPE has better Young's Modulus value in comparison with PALF-LDPE. Among these three composites, PALF-LDPE system appeared to have the

highest elongation at break values. Actually PALF-LDPE has indicated these superior performances due to the high cellulose content.

Table 2.6: Comparison of Tensile Properties of Randomly Oriented PALF-LDPE, Sisal Fibre-LDPE and Jute Fibre-LDPE Composites [a] (George *et al.*, 1995)

	Tensile Strength (MP'a) Fibre Loading			**Young's Modulus (MPa)** Fibre Loading			**Elongation at Break (%)** Fibre Loading		
Fibre	10	20	30	10	20	30	10	20	30
PALF	10.2	11.4	13.0	218	366	570	24	12	6
	(16.3)	(19.8)	(22.5)	(610)	(900)	(1100)	(11)	(9)	(4)
Sisal	10.8	12.5	14.7	324	453	781	2	1	
	(15.6)	(21.0)	(31.0)	(1429)	(2008)	(3086)	(4)	(3)	(1.8)
Jute	-	4.99	8.03	-	-	-	-	11.8	4.9

[a] Values in parentheses correspond to longitudinally oriented composites

2.6 Flexural Properties of PALF Composite

Although there are not many test carried out for the flexural properties of composites, this properties is important to study the fracture behavior. Regarding PALF reinforced polyester, Uma Devi *et al.* (1997) pointed that the addition of fibre made the composite more ductile. In his work, the flexural strength values of the PALF-polyester composites were less than the pure polyester at low weight fractions (10 wt %) of the fibre. Further increased of the fibre loading, the flexural strength has improved by nearly 120 % for composite containing 30mm long fibres. Figure 2.7 is the representation of these arguments. But when reach more than 30 wt %, the flexural modulus decreased. At 40 wt %, the flexural strength decreased cited 13 % of value. To explain this, the author pointed that higher fibre loading

encouraged fibre-to-fibre interaction and the fibre were not well dispersed within the resin latex. For this research, the best result was obtained at fibre loading of 30 wt %.

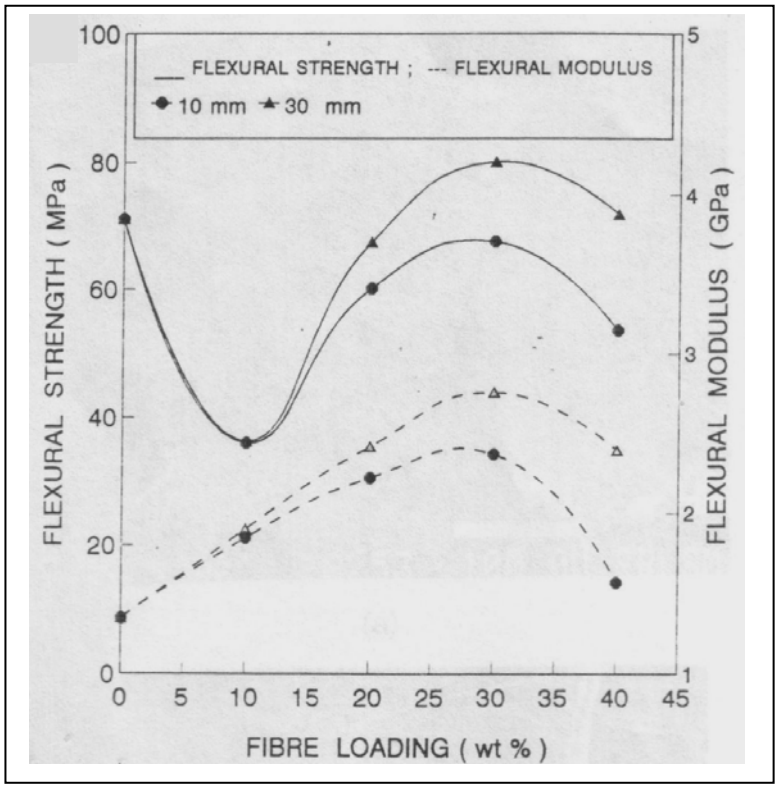


Figure 2.8: Variation of Flexural Strength and Flexural Modulus with Fibre Loading of PALF-Polyester Composites (Uma Devi *et al.*, 1997)

2.7 Impact Properties of PALF Composite

Impact properties have been studied for PALF-polyester by (Uma Devi *et al.*, 1997). They focused on the work of fracture and impact strength of PALF composites as a function of fibre loading. The impact strength increased almost linearly with the weight fraction of the fibre as shown in Figure 2.9. For a 30 wt %

of fibre, the impact strength of composite is found to be 24 kJ m^{-2}. This was roughly 1200 % greater than that of pure polyester resin. Typically, most people accepted that the toughness of a fibre composite is depending on the fibre stress-strain behavior. For the fact that PALF are comparatively strong fibres with high failure strain, it imparts a high work of fracture to the composite. Fibre pullout and interface fracture were the major contributions toward the high toughness of these composites.

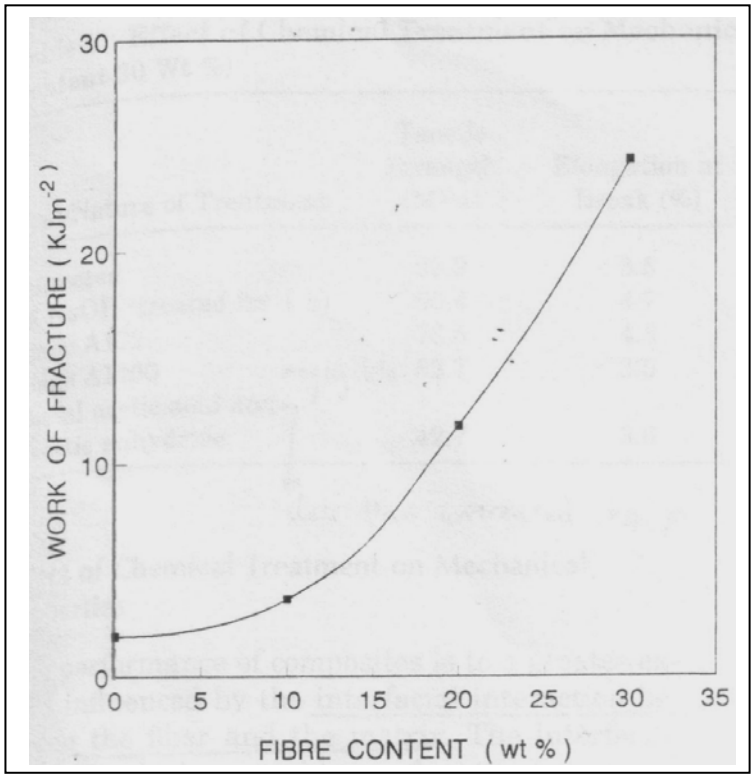


Figure 2.9: Variation of Work of Fracture of PALF-Polyester Composites with Fibre Content (Fibre Length 30mm) (Uma Devi *et al.*, 1997)

Pavithran *et al.* (1987) comparatively studied on the of impact properties of unidirectional aligned polyester reinforced with sisal, pineapple, banana and coir fibre. The work of fracture was found maximum in sisal composites yielding toughness of 98.7 kJm^{-2}. This was followed by PALF composite with toughness achieved of 79.5 kJm^{-2}. The PALF composites exhibited high toughness than banana leaf fibre was likely to arisen up out of the complete fracture behavior of the composites. As for the low toughness in coir composites, it was due to the highly viscoelastic nature of the fibre. This led to failure in matrix first then followed by the

fracture of fibre at the crack plane. The effect of microfibrillar angel was observed and related to the work of fracture as well.

CHAPTER 3

MATERIALS AND METHODOLOGY

3.1 Introduction

This chapter is an amalgamation of three basic stages:

- Raw material preparation
- PALF Characterization
- Sample preparation
- Testing

3.2 Raw Materials Preparation

3.2.1 Polypropylene

Polypropylene that was used as matrix for PALF reinforced composite is a homo polypropylene TITANPRO 6331 supplied by Titan PP Polymers (M) Sdn. Bhd.

This polymer is for general purpose of injection molding. Its properties are summarized in Table 3.1.

Table 3.1: Properties of Polypropylene (TITANPRO 6331)

Typical Resin Properties	**Values**	**ASTM Methods**
Melt Flow Rate, at 230^{o}C (g/10min)	14	D1238
Density (g/cm^{3})	0.9	D1505
Tensile Strength at Yield (kg/cm^{2})	360	D638
Elongation at Yield (%)	10	D638
Flexural Modulus (kg/cm^{2})	17500	D790B
Notched Izod Impact Strength at 23^{o}C (kg.cm/cm)	2.6	D256A
Heat Deflection Temperature at 4.6 kg/cm^{2} (^{o}C)	99	D648
Rockwell Hardness (R scale)	97	D785A
Water absorption after 24 hours (%)	0.02	D570

Note:
- The typical average values and not specification.
- Sources from http://www.titangroup.com/Product/TitanPro/HTML/Pro6331.htm (Appendix A)

3.2.2 Preparation of PALF

PALF is contained in the spiky leaves of pineapples. The waste pineapples leaves were collected from Sedenak or Pekan Nanas, Johore during the harvest process. These leaves were pressed using two-roll mill to remove circa 90 % of the water content. Figure 3.1 shows the Rapic Two-roll Mill machine that was utilized. Fibres were extracted manually from these semi-dried leaves by knife or sharp edge tool. Figure 3.2 indicates the original fibre extracted from leaf and the chopped PALF. They were then chopped to particle sizes of 2-6 mm. This particular size

appears to be the most suitable processing size for melt mixing which give effective reinforcement in PP (George *et al.*, 1995). The short fibres were then undergone cleaning steps where they were washed thoroughly in 2 % detergent solution at 70˚C followed by tape water. Basically, this removed most of the foreign objects and impurities inside the fibres. They were later dried in oven at 70˚C for 24 hours before characterizing and chemical treatment for further processing.

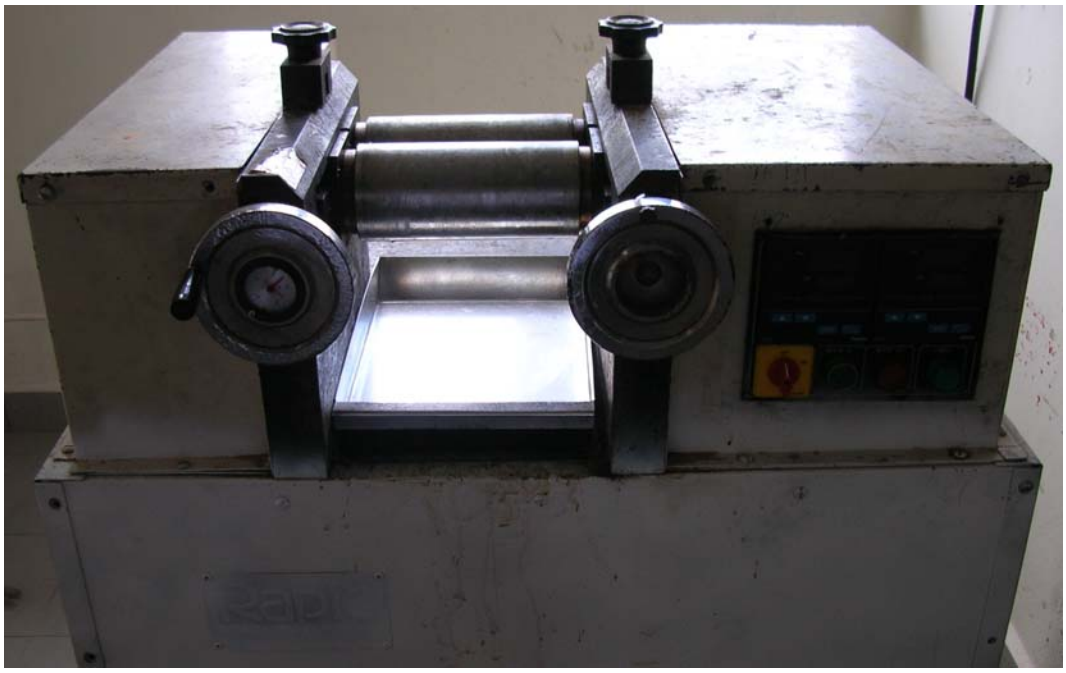


Figure 3.1: Rapic Two-roll Mill Machine Used for Remove Water and Composite Compounding

(a)

(b)

Figure 3.2: PALF Photos: (a) Long PALF Extracted from Raw Leaf and (b) Chopped PALF

Prior to composite preparation, the fibres were treated with coupling or chemical agents such as silane, isocyanate, alkali and peroxide to improve the adhesion between the matrix and fibre. In this research, alkaline group of sodium hydroxide (NaOH) was used. Fibre was treated with 5 % NaOH solution for one hour at 30˚C. This alkali treatment removed natural and artificial impurities which improves the fibre-matrix adhesion. It may also destroy the hydrogen bonding in cellulose hydroxyl groups of the fibre, thereby making them more reactive to the functional group of coupling agent, which in turn bonds to the polymer matrix. Good fibre – matrix bonding can be established (Mishra *et al.*, 2001; George *et al.*, 1998). The fibres after air dried were ready to serve as reinforcing agents in matrix. Since PALF are susceptible to water content, the thoroughly drying and storing in a dry condition are crucial. Silica gel was placed in the desiccator together with PALF to prevent any water absorption.

3.3 PALF Characterisation

3.3.1 Extraction (Methanol-Toluene Solubility)

This method describes a procedure for extraction of non-wood fiber for further analysis, such as holocellulose, hemicellulose , cellulose and lignin analysis.

- The test was carried out in duplicate
- 2-3g of PALF samples were weighted into a thimble
- The thimble was placed into the Soxhlet unit
- 200ml of the Methanol-Toluene [2:1] was placed in a 500ml round bottom flask
- The extraction was carried out for 6-8hrs

- It was then dried to constant weight in oven at 105^{o}C
- It was cooled in the desiccator
- The sample was again weighted until the constant weight

3.3.2 Preparation of Holocellulose (Chlorite Holocellulose)

Holocellulose is defined as a water-insoluble carbohydrate fraction of plant materials. The sample should be extractive and moisture free and prepared after procedure 3.3.1.

- The extracted residue from Methanol-Toluene solubility was used
- The residue was then transferred to 250ml tall beaker
- To 2.5g of sample, 80ml of distilled water, 0.5ml acetic acid, and 1g of sodium chlorite were added
- The mixture was heated on a water bath at 70^{o}C
- After 60min, 0.5ml acetic acid and 1g sodium chlorite were added
- After each succeeding hour, fresh portions of 0.5ml acetic acid and 1g sodium chlorite were added with shaking
- At the end of 24hrs of reaction, the sample was cooled and filtered the holocellulose on filter paper until the yellow color (holocellulose is white) and the odor of chlorine dioxide were removed
- Finally washed with acetone
- Vacuum-oven dried at 105^{o}C for 24hrs
- Transfer to a desiccator
- Weigh at daily intervals until the sample reaches constant weight

The holocellulose should not contain any lignin and the lignin content of holocellulose should be determined and subtracted from the weight of the prepared holocellulose.

3.3.3 Preparation of α-Cellulose (Determination of Hemicellulose)

Extractive-free, lignin-free holocellulose was treated with sodium hydroxide and then with acetic acid, with the residue defined as α-Cellulose. Thus the last fraction gives the hemicellulose content.

- 2g of vacuum-oven dried holocellulose was weighted and placed into a 250ml glass beaker
- 10ml of 17.5% NaOH solution was added (manipulate the holocellulose lightly with glass rod so that all of the specimen becomes soaked with the NaOH solution)
- 5ml more of the NaOH solution was added and thoroughly stirred the mixture with the glass rod, until the NaOH was gone [repeated 3 times, at 5 minute intervals]
- Allowed to stand at 25^0C for 30 minutes (total time for NaOH treatment was 45 minutes)
- 33ml of distilled water was added at 25^oC to the mixture. The contents of the beaker was thoroughly mixed and allowed to stand at 25^oC for 1hr before filtering
- The cellulose was filtered with the aid of suction into a tarred, alkali-resistant Alundum or fritted-glass crucible of medium porosity
- The holocelluse residue was transferred on the crucible, washed it with 100ml of 8.3% NaOH solution at 25^oC, continued the washing with distilled water, disconnect suction, filled the crucible to within 6 mm of the top with water, carefully breaking up the cellulose mat with a glass rod to separate any lumps present, and again applied suction. Repeated this step twice.
- Filled crucible with 15ml of acetic acid (room temperature)
- Allowed to stand for 3 minutes
- Reapplied suction to remove acetic acid
- Washed free of acid with distilled water, as indicated by litmus paper

Calculation and report

The percentage of α-Cellulose on the basis of the oven-dry holocellulose sample was calculated as follows:

α-Cellulose, percent = (W2/W1) x 100

W2 = weight of the oven-dry α-Cellulose residue

W1 = weight of the original oven-dry holocellulose sample

3.3.4 Preparation of Klason Lignin

- Sample were prepared by extraction (Methanol-Toluene Solubility)
- The sample was dried at 45^{o}C in a vacuum-oven overnight
- 200mg of vacuum-oven dried sample was weighted into a 100ml centrifuge tube
- 1ml of 72%(w/w) H_2SO_4 sulfuric acid for each 100mg of sample was added
- The mixture was stirred and dispersed thoroughly with glass rod twice, then incubated the tubes in a water bath at 30^{o}C for 1 hr
- 56ml of distilled water (use 60ml syringe) was added
- Autoclaved at 121^{o}C, 15psi, for 60 min.
- The samples was removed from the autoclave and filtered off the lignin, with glass fiber filters (filters were rinsed into crucibles, dried and tarred) in crucibles using, keeping the solution hot.
- The residue was washed thoroughly with hot water and dried at 105^{o}C overnight.
- It was then moved to a desiccator and let it sit 1hr and weighed to five places.
- Calculated Klason lignin content from weights

3.4 Sample Preparation

The preparation of sample was carried out in two stages, namely PALF-PP composite preparation and testing sample preparation.

3.4.1 PALF-PP Composite Preparation

Melt mixing method was used to mix PALF and PP. Various compositions of PALF in PP were compounded in two-roll mill as shown in Figure 1. Table 3.2 indicates different fibre loadings that are presented in the PP matrix. Coupling agent - Compatibilizer Licomont AR 504 supplied by Clariant was added into the

compound to improve the durability and mechanical strength of the interface region between PALF and PP matrix. Its material data sheet is attached in Appendix B. Figure 3.3 shows the natrium hidroxide used in chemical treatment and coupling agent.

Table 3.2: PALF Reinforced PP Composites Composition

Ingredient	Composition (%wt)				
	S1	**S2**	**S3**	**S4**	**S5**
Polypropylene (PP)	100	90	80	70	60
Pineapple Leaf Fibre (PALF)	0	10	20	30	40
Coupling agent*	1	1	1	1	1

* Coupling agent was added in terms of phr.

Figure 3.3: Left is the Natrium Hydroxide Used for Chemical Treatment while the Right is the Coupling Agent – Licomont AR504 Used

Composition of PP and PALF were weighted accordingly and melt mixed for a period of 30 minutes at temperature of 180˚C using Rapic two-roll mill. The temperature was used as the optimum processing temperature for PP matrix but not exceeding that. Higher temperature would result in a reduction in strength and

modulus which may be due to the degradation of fibre. In addition, dispersion of PALF in PP matrix will be poor as there is a drop in viscosity at high temperature. The compounded composites were thermoformed by hot press machine with thickness of 1 mm and 3 mm according to the specs that are required in testing sample. The operating temperature was 190˚C with 15 minutes of preheat, 10minutes of compression and 12 minutes of cooling. Figure 3.4 (a) shows the hot press machine whereas Figure 3.4 (b) shows cooling machine for cooling session. Preheat was needed to melt the scattered compound from two-roll mill and to promote flow of resin to every hill and valley of the fibre. In consequent, there was no significant resin reached or voids introduced. Last but not least, the thin plates of composites were trimmed and stored with silica gel. Figure 3.5 is the laminate of composite produced via hot press.

(a) (b)

Figure 3.4: Machineries: (a) Hot Press Machine and (b) Cooling Machine

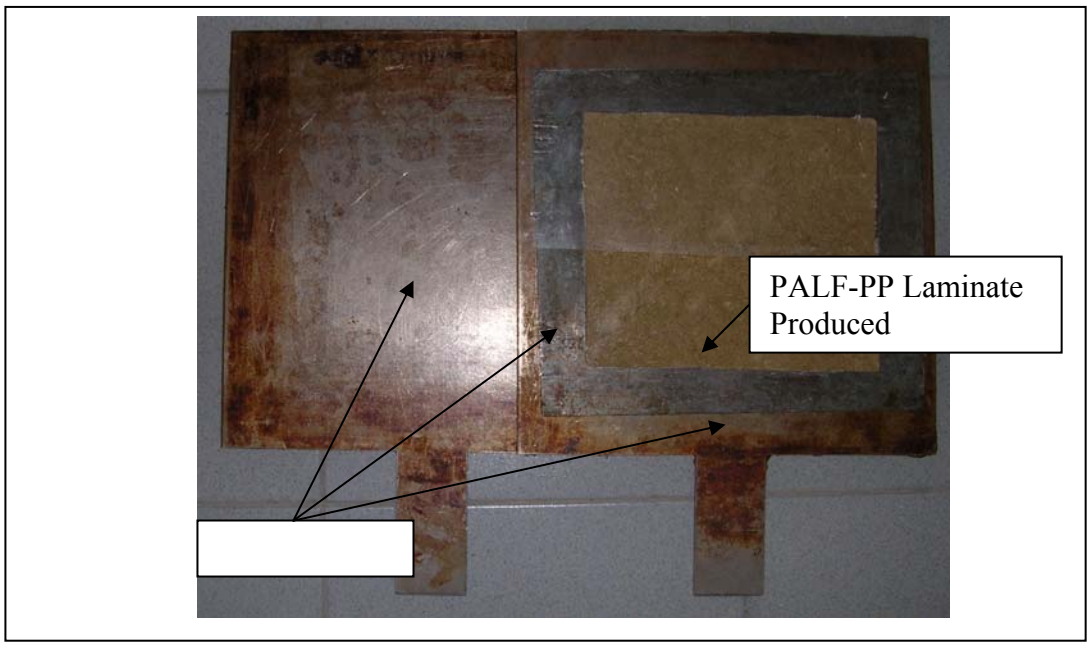


Figure 3.5: PALF Reinforced PP Laminate Produced Via Hot Press Machine

3.4.2 Testing Sample Preparation

PALF-PP composite were hot pressed into 1 mm thin plate for tensile specimens and 3 mm thickness for both flexural and impact test specimens. All these specimens were machined into shape using grinding machine according to standards discussed later. Figure 3.6 is a photo of grinding machine with one example of tensile speciment's specification.

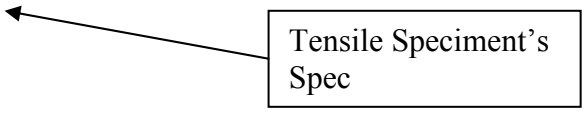


Figure 3.6: Grinding Machine and Tensile Specimen's Specification

3.5 Testing Methods

All the mechanical testing methods that were carried out were base on American Standard Testing Methods (ASTM). There were three test performed, namely Tensile Test (ASTM D638), Flexural Test (ASTM D256) and Impact Test (ASTM D790). For morphology studies, Scanning Electron Microscope (SEM) was used.

3.5.1 Tensile Testing

In a broad sense, tensile test is a measurement of the ability of a material to withstand forces that tend to pull it apart and to what extent the material stretches before breaking. The stiffness of a material which represented by tensile modulus can be determined from stress-strain diagram.

According to ASTM (D638), dumbbell shape (Type I) specimen is needed for reinforced composite testing. Detail dimension for this are shown in the Figure 3.7 and Table 3.3. The testing were done in standard laboratory atmosphere of 23˚C ± 2˚C (73.4˚F ± 3.6˚F) and 50 ± 5 percent relative humidity. This condition of plastic for not less than 40 hours prior to test in accordance with Procedure A of ASTM D618. Universal Testing Machine (Instron 5567) was used at cross-head speed of 50 mm/minute. Figure 3.8 shows the Universal Testing Machine (Instron 5567) used for PALF-PP tensile testing. The specimens were positioned vertically in the grips of the testing machine. The grips were then tightened evenly and firmly to prevent any slippage with gauge length kept at 50mm. The precise five tested result were chosen for each fibre loading of PALF in PP matrix.

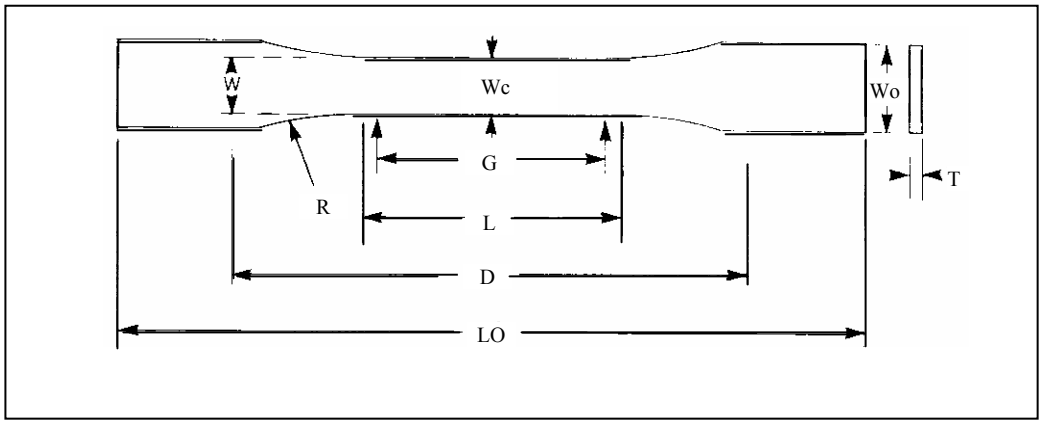


Figure 3.7: Dumbbell Shaped Specimen Dimension for Type I in ASTM D638

Table 3.3: Dumbbell Shaped Specimen Dimension for Type I in ASTM D638

Dimension	Value, mm (in)
Thickness <7mm (0.28in), T	1.00 ± 0.4 (0.13 ± 0.02)
Width of narrow selection, W	13 (0.5)
Length of narrow selection, L	57 (2.25)
Width overall, WO	19 (0.75)
Length overall, LO	165 (6.5)
Gauge length, G	50 (2.00)
Distance between grips, D	115 (4.5)
Radius of fillet, R	76 (3.00)

Figure 3.7: Universal Testing Machine - UTM (Instron 5567) for Tensile and Flexural Testing

As the tensile test starts, the specimen elongates; the resistance of the specimen increases and is detected by a load cell. This load value (F) is recorded until a rupture of the specimen occurred. Instrument software provided along with the equipment will calculate the tensile properties for instance tensile strength, yield strength and elongation at break. Below are the basic relationships to determine these properties:

$$\textit{Tensile strength} = \frac{\textit{Force (load)}}{\textit{Cross section area}} \quad \text{------ (3.1)}$$

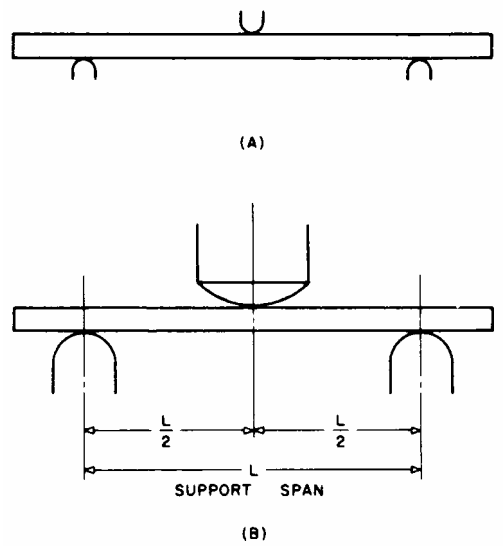
$$\textit{Tensile strength at yield} = \frac{\textit{Maximum load recorded}}{\textit{Cross section area}} \quad \text{------ (3.2)}$$

$$\textit{Tensile strength at break} = \frac{\textit{Load recorded at break}}{\textit{Cross section area}} \quad \text{----- (3.3)}$$

3.5.2 Flexural Testing

Flexural strength is the ability of the material to withstand bending forces applied perpendicular to its longitudinal axis. Sometime it is referred as cross-breaking strength where maximum stress developed when a bar-shaped test piece, acting as a simple beam, is subjected to a bending force perpendicular to the bar. This stress decreased due to the flexural load is a combination of compressive and tensile stresses. There are two methods that cover the determination of flexural properties of material: three-point loading system and four point loading system. As described in ASTM D790, three-point loading system applied on a supported beam was utilized. Flexural test is important for designer as well as manufacturer in the form of a beam. If the service failure is significant in bending, flexural test is more relevant for design and specification purpose than tensile test.

According to ASTM D790, specimens of test pieces were prepared with dimension of 127mm × 12.7mm × 3.2mm (5 in × ½ in × ⅛ in). The test pieces were tested flat wise on a support span resulting span-to-depth ratio of 16. This means the span is 16 times greater the thickness of specimen. In Procedure A of ASTM D790, width and depth of the specimen were measured to the nearest 0.03mm (0.001 in) at the centre of the support span. The test pieces were then placed on two supports and load will be applied. The distance of two supports span (L) was fixed at 100mm. Figure 3.8 shows the requirement of loading nose and support radii.



(a) Minimum radius = 3.2 mm (⅛ in).
(b) Maximum radius support 1.6 times specimen depth; maximum radius loading nose = 4 times specimen depth

Figure 3.8: Allowable Range of Loading Nose and Support Radii in ASTM D790

Flexural test was done by Universal Tensile Machine (Instron 5567) at standard laboratory atmosphere of 23˚C ± 2˚C (73.4˚F ± 3.6˚F) and 50 ± 5 percent relative humidity. The load applied at specified cross-head rate was fixed for a value within the ±10% of the calculated R using equation (3.4). (Please refer Appendix C for calculation steps). This constant cross-head motion appeared to be 5mm/min.

$$R = ZL^2/6d \qquad \text{----- (3.4)}$$

where R = rate of cross head motion, mm/min [in/min]

Z = rate of straining of the outer fibre, mm/mm/min [in/in/min] = 0.01

L = support span, mm [in]

d = depth of beam, mm [in]

The constant load was then applied on test piece and deflection is recorded. The testing will be terminated when the maximum strain in the outer surface of the specimen has reached the maximum strain of 5 % or rupture occurs. Five consistent test pieces results were chosen for each fibre loading composition.

There were two important parameters being determined in the flexural test, they are flexural strength and tangent modulus of elasticity in bending.

(i) Flexural Strength

Flexural strength is the maximum stress in the outer specimen at the moment of break. When the homogeneous elastic material is tested with three-point system, the maximum stress occurs at the midpoint. This stress can be evaluated for any point on the load deflection curve using equation (3.5).

$$\sigma_f = \frac{3PL}{2bd^2} \qquad \text{----- (3.5)}$$

where σ_f = stress in the outer specimen at midpoint, MPa [psi]

P = load at a given point on the load deflection curve, N [lb_f]

L = support span, mm [in]

b = width of beam tested, mm [in]

d = depth of beam tested, mm [in]

(ii) Tangent Modulus of Elasticity

Modulus of elasticity or flexural modulus is a measure of the stiffness during the initial of the bending process. This tangent modulus is the ratio within the elastic limit of stress to corresponding strain. A tangent line will be drawn to the steepest initial straight line portion of the load deflection curve and the value can be calculated using equation (3.6).

$$E_B = \frac{L^3 m}{4bd^3} \qquad \text{----- (3.6)}$$

where E_B = modulus of elasticity in bending, MPa [psi]

L = support span, mm [in]

m = slope of the tangent to the initial straight line portion of the load-deflection curve, N/mm [lb_f/in] of deflection

b = width of beam tested, mm [in]

d = depth of beam tested, mm [in]

3.5.3 Impact Testing

The impact properties of the material are directly related to the overall toughness which is defined as the ability to absorb applied energy. Area under the stress-strain curve is proportional to the toughness of a material. Nevertheless, impact strength is a measure of toughness. In this last two decades, there are four types of impact tests, for example: the pendulum impact tests, high-rate tension test, falling weight impact test and instrumented impact test. In this research, pendulum impact test – Notched Izod Impact Test was utilized as shown in Figure 3.9.

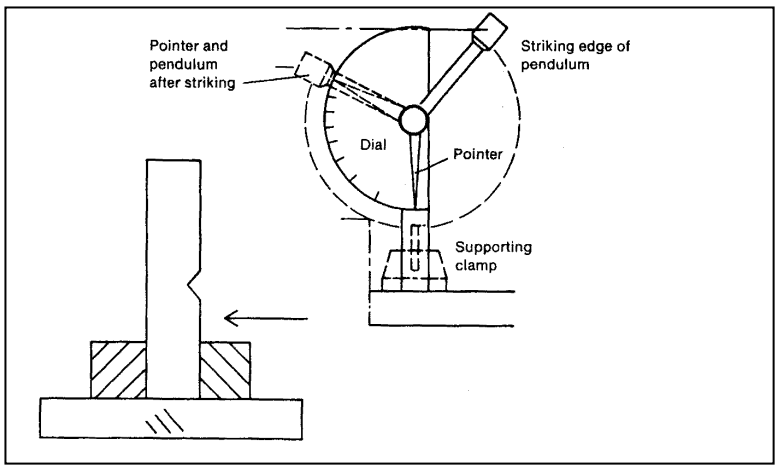


Figure 3.9: Pendulum Impact Test – Notched Izod Impact Test

According to ASTM D256, test method A (Izod type) was used for testing. The apparatus involved was Cantilever Beam (Izod Type) Impact Machine and the specimens were notched. Notching was done because it provides a stress concentration area that promotes a brittle rather than a ductile failure. Furthermore, notching also drastically reduces the energy loss due to the deformation of plastic.

In the testing, specimens were clamped vertically as a cantilever beam and then struck by a single swing of the pendulum released from a fix distance from the specimen clamp. The line of initial contact is at a fixed distance from the specimen

clamp and from the centerline of the notch and on the same face of the notch. Figure 3.10 shows the relation of vise, specimen and striking edge to each other for Izod Test and Figure 3.11 illustrates the specimen dimension. A photo of impact speciments is shown in Figure 3.12. Total of five consistent testing results were selected for each fibre loading in PP matrix. In this work of research, RAY-*RAN* Universal Pendulum Impact System for Izod-Charpy-Tension and Puncture was used to measure the work of fracture for PALF-PP composite. This equipment is shown in Figure 3.13. There are a few parameters that are set according to the standard for instances, Hammer Velocity = 3.46 m/s and Hammer Weight = 0.905 kg.

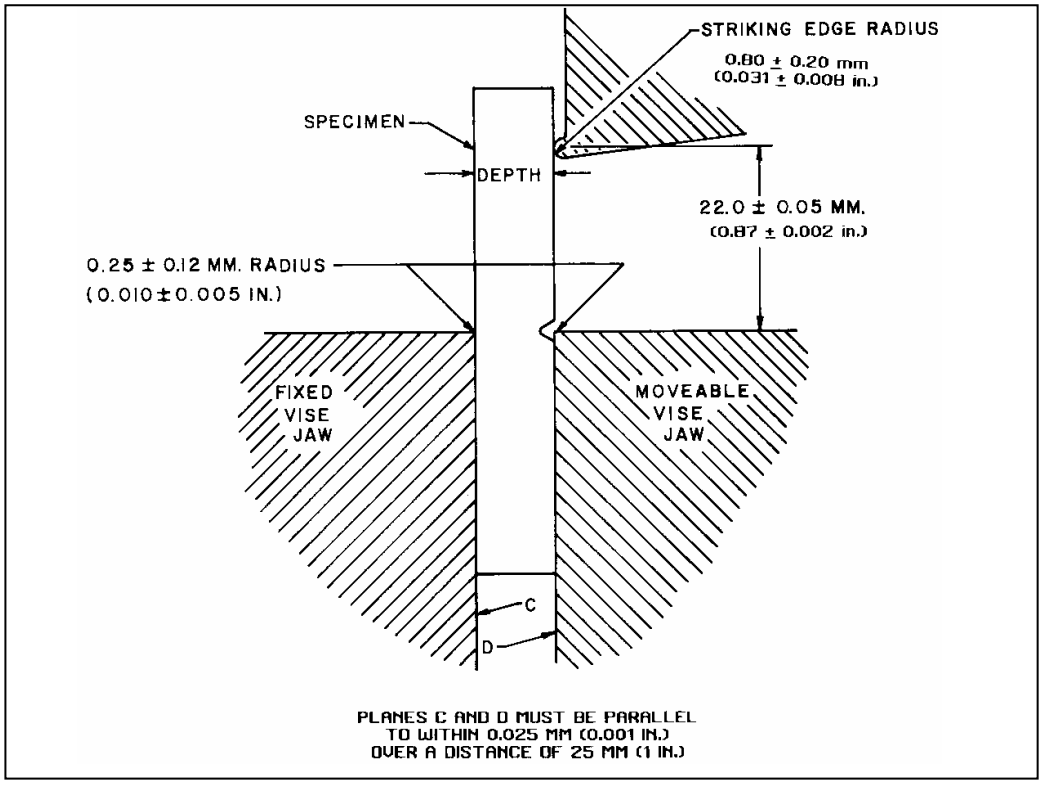


Figure 3.10: Relation of Vise, Specimen and Striking Edge to Each Other for Izod Test Method A in ASTM D256

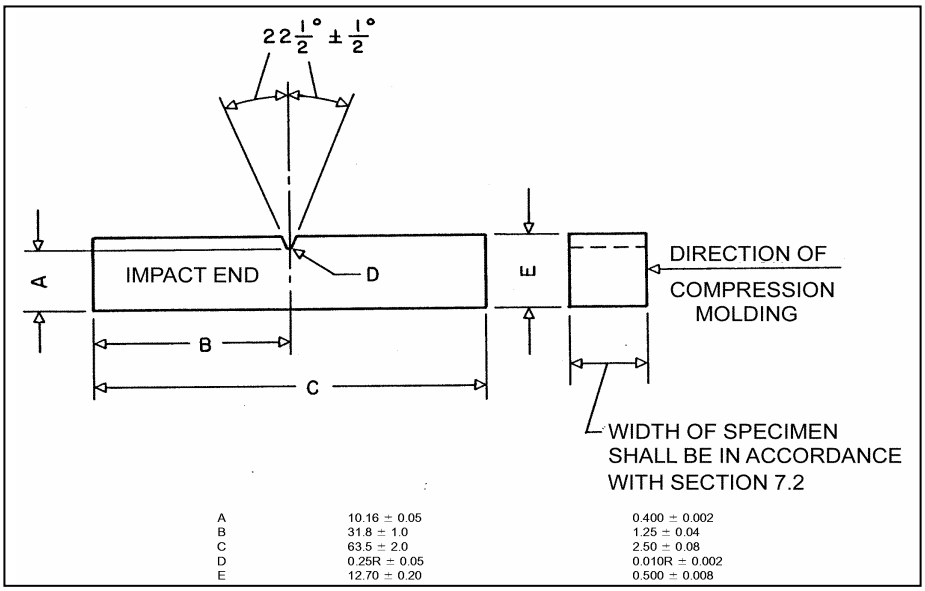


Figure 3.11: Dimension of Izod Test Specimen in ASTM D256

Figure 3.12: Impact Testing Specimens for Various wt% of PALF Reinforced in PP

Figure 3.13: RAY-*RAN* Universal Pendulum Impact System for Izod-Charpy-Tension and Puncture

3.5.4 Scanning Electron Microscope (SEM)

For morphological study, Scanning Electron Microscope (SEM) was used to reveal the fibre orientation in reinforced thermoplastics together with some information concerning the nature of the bond between the fibres and matrix. It is an instrument for obtaining micro structural images using a scanning electron beam. In the SEM, a small electron beam spot (usually circa 1μm) is scanned repeatedly over the surface area of the sample. The importance of SEM is it produces image that likes a visual image of a large scale piece which allows the irregular surface of the material to be observed.

In this research, the SEM was used to study the miscibility and interactions between PALF and PP. SEM poses a few advantages, such as more realistic images (in the form of three dimensional), deep focusing, easy to use and ease of specimen preparation. The micrographs produced showed surface topography and chemical contrast at the shortest time. JEOL JSM-5600LV Scanning Electron Microscope was used. The photo of this equipment is shown in Figure 3.14. Before performing SEM, PALF-PP composites were dipped into liquid nitrogen to promote and ease brittle fracture of the sample. Secondly, the samples were discharged and broke into two portions. The specimens were placed on a stub, coated with platinum and inserted into the scanning barrel. The inter condition of the scanning barrel were vacuumed to prevent interference of scanning picture due to the presence of air. Magnification, focus, contrast and brightness of the result were adjusted to produce the best micrographs.

Figure 3.14: JEOL JSM-5600LV Scanning Electron Microscope

CHAPTER 4

RESULT AND DISCUSSIONS

4.1 Introduction

This chapter covers characterization of PALF and mechanical properties of the short treated PALF reinforced PP and its morphological analysis.

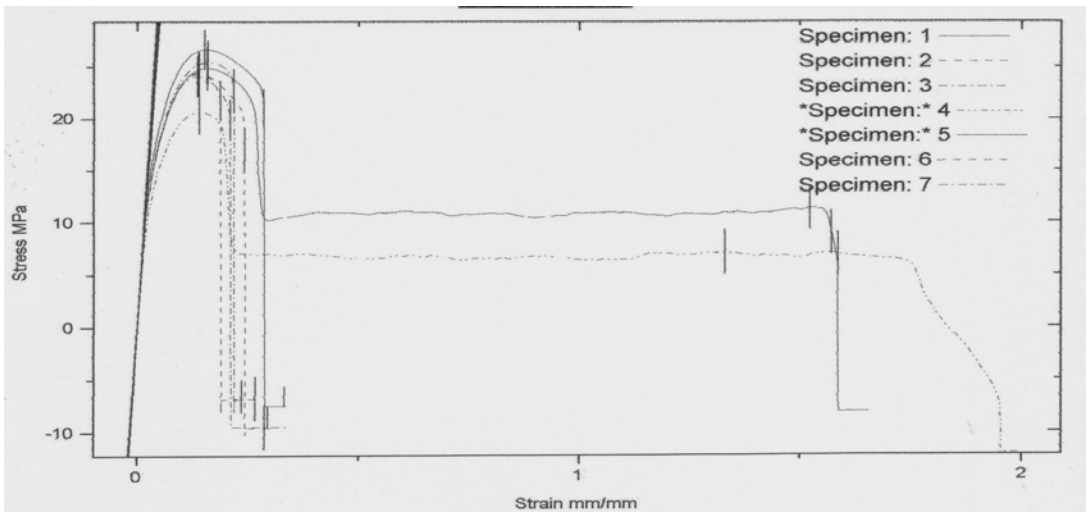
4.2 Characterization of PALF

From the experiments, it was found that PALF contains the following chemical constituents:

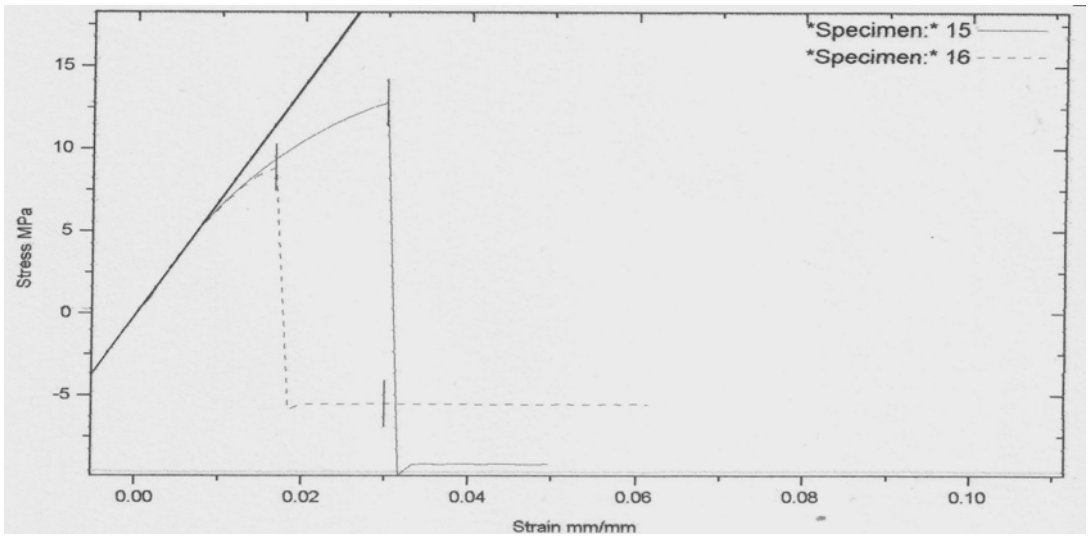
Chemical Constituents	% Composition
Holocellulose	87.56
Alpha- cellulose	78.11
Hemicellulose	9.45
Lignin	4.78

4.3 Tensile Properties

Figure 4.1 indicates the typical stress-strain diagram of unreinforced PP and treated PALF reinforced PP tested at crosshead speed of 50 mm/min. After the initial linear region (elastic behavior), the curvature of reinforced composite observed was not significant compared to the unreinforced PP. The composite finally failed with further increase in stress like brittle material. In comparison, unreinforced PP exhibited necking where the polymer molecule would highly oriented and then fibrillation occured. This is phenomenon wherein polymer showed further evidence of basic fibrous structure or fibrillar crystalline nature, by a longitudinal opening-up under rapid, excessive tensile or shearing stresses. Incorporation of random short fibre has interrupted the necking behavior as they acted as foreigners to absorb stress to certain amount.



(a)



(b)

Figure 4.1: Typical Stress-strain Curves for (a) Unreinforced PP and (b) Short treated PALF Reinforced PP Composite

In tensile test, the most properties can be represented by Young's modulus and tensile strength. Figure 4.2 shows that Young's Modulus increased with fibre loading at the beginning and experienced drastic raise at 30 wt % of fibre loading, that was from 649 MPa to 997 MPa. After 30 wt % fibre loading, the modulus dropped to almost the same value in 10 wt% fibre loading, which was 667 MPa. To be more precise, the addition of 10 wt % PALF did not significantly increase the modulus but slowly with 20 wt % PALF, the modulus showed an increment of 10% whereas for 30 wt % PALF, the modulus was 53% higher than the unreinforced PP (0 wt% PALF). From observation, the optimum fibre loading which yield the highest Young's modulus was at 30 wt %. These increased of modulus actually mean that the PPs with reinforcement were becoming stiff and could withstand higher stress at the same strain portion. The fibre served as reinforcement because the major share of load has been taken up by the crystalline fibrils resulting in extension of the helically wound fibrils along with the matrix (Mukherjee and Satyanarayana, 1986). For 40 wt %, there was drop in modulus. This is because at higher volume fraction of fibre, the fibre acted as flaws and crazing occured, thus creating stress concentration area which lowering the stiffness of composite. Besides, the 40 wt % of fibre was a bit excessive that the PP matrix was hard enough to flow through every fibre thus leaving voids and fibres are more easily expose to environmental degradation. In practical, this composition is hard to produce and the composite is brittle.

On the same graph, for tensile strength, it was decreasing as the fibre loading increased in overall. However there was a plateau of tensile strength after 10 wt % until 30 wt% fibre loading and after this optimum 30 wt%, the value experienced a critical drop. Actually, the results obtained slightly deviates from the line graph drawn. This may due to the incompetence of tensile test piece preparation; this includes non-uniform specimen specification in width, thickness and gauge length. The result is still acceptable because the standard deviation obtained is still

acceptable. To conclude, these ultimate stresses before break basically decreased because the interfacial adhesion between fibre and PP was not good, fibre-fibre interaction was preferred by the system. Fibre agglomerations happened thus causing dispersion problems in PP, which lead to decrement in tensile strength (George *et al*., 1995).

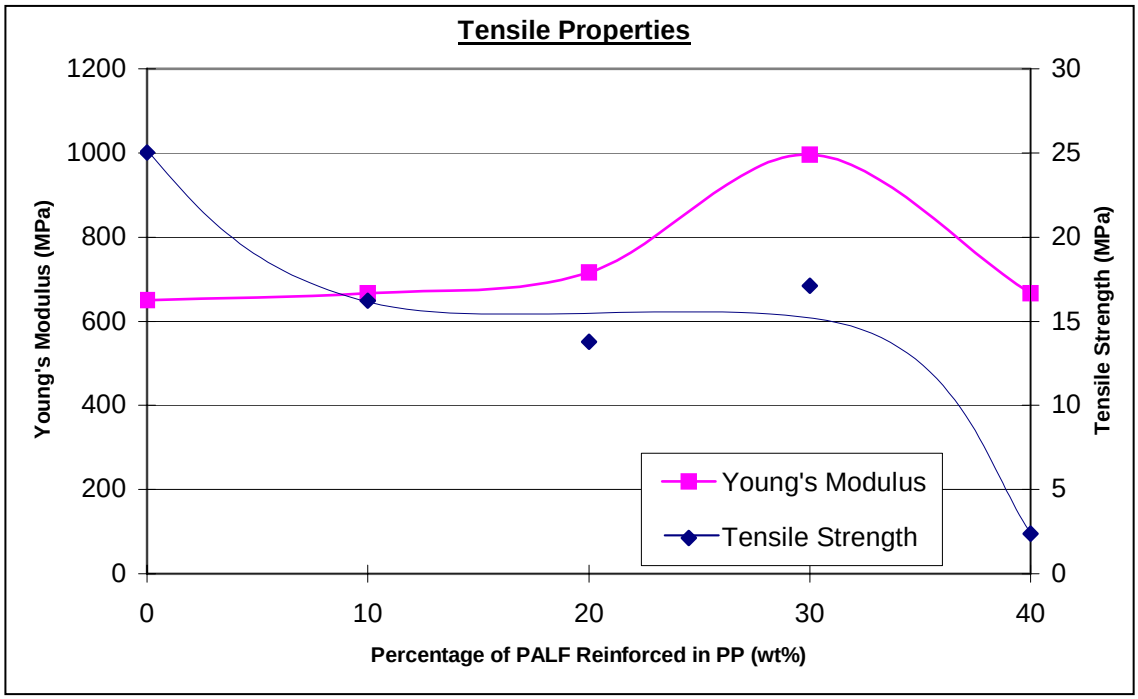


Figure 4.2: Tensile Properties of Short treated PALF Reinforced in PP

On the other hand, elongation at break of the unreinforced and PALF reinforced PP are shown in Figure 4.3. A decreasing trend in elongation at break was demonstrated with the increment of fibre loading. The histogram shows that the decrement was not stable with a slight higher elongation at break for fibre composition of 30 wt %. However the standard deviation obtained was still acceptable and the changed in elongation at break after 10 wt% PALF were not significant (less than 5 %). As a result, a modified smooth trend line is drawn to explain the more realistic case. The elongation at break experienced significant drop at 10 wt% PALF and maintained almost linearly as the fibre composition increased. This phenomenon observed because the addition of stiff fibre again interrupted the PP segment mobility and thus turning the plastic to be more brittle.

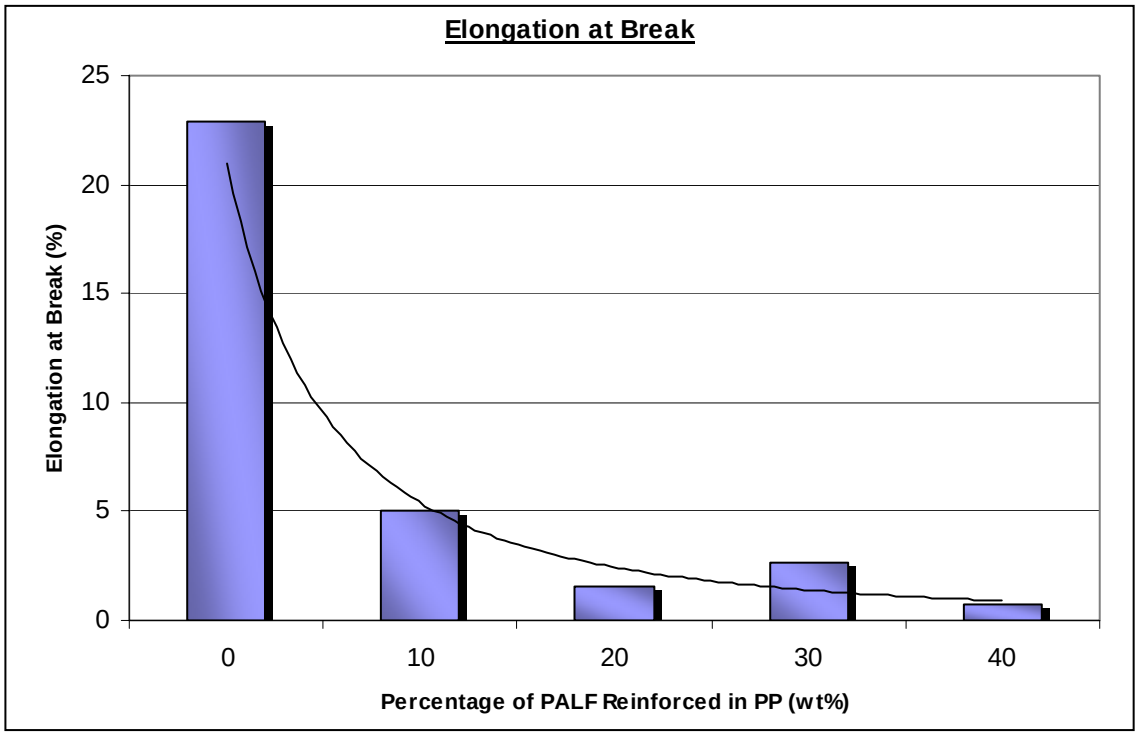


Figure 4.3: Graph of Elongation at Break for Varies Short PALF Composition in PP

Table 4.1 is the comparison of tensile properties for PP which reinforced with short and long PALF. From birds eye view, tensile strength as well as elongation at break for long fibre is generally more dominant than short fibre. The fibre ends of short PALF are normally weak points in the composite site of high stress concentration in the matrix and meanwhile, continuous long PALF extends the entire length of the specimen, so that there are no weak spot occur. This has contributed to the higher properties of long fibre in tensile strength and elongation at break compare to short fibre.

Table 4.1: Tensile Properties of Long and Short treated PALF Reinforced PP with Various Fibre Composition

	Long PALF-PP*		**Short treated PALF-PP**	
Fibre Content (wt%)	Tensile Strength (MPa)	Elongation at Break (%)	Tensile Strength (MPa)	Elongation at Break (mm/mm)
0	35.1	33.9	25.0	22.9
10	36.2	21.2	16.2	5.0
20	33.9	16.1	15.5	1.6
30	34.9	11.8	15.3	2.7
40	31.6	9.8	2.4	0.7

* These results obtained from Arib (2003).

4.4 Flexural Properties

Flexural strength and flexural modulus for PP and PP composite are shown in Figure 4.4. From this figure, both flexural strength and flexural modulus were increasing gradually with fibre loading. It is observed that the flexural strength increased from 22.8 MPa to 38.9 MPa and flexural modulus increased from 1392 MPa to 2945 MPa respectively for pure PP to 40 wt% fibre. The addition of 40 wt % fibre has obviously increased the flexural strength and flexural modulus of unreinforced PP as much as 112 % and 70 %. The flexural strength of the composite increased linearly with fibre composition and it was significantly higher than corresponding tensile strength obtained in experiment. This is to say that PALF reinforced PP can withstand bending forces better than tensile stress. Table 4.2 is a summary of both tensile strength and flexural strength with various fibre loadings. All these observations are to prove that PALF which has high crystalline content are strong and can share the load applied in matrix effectively with crystalline fibrils in it. Mukherjee and Satyanarayana (1986) again highlighted that the PALF in the

composite system will defect and fail when the stress initiated the defective cells as a result of stress concentration. In consequently, the PALF can withstand bending forces which comprise of compressive forces and tensile stress.

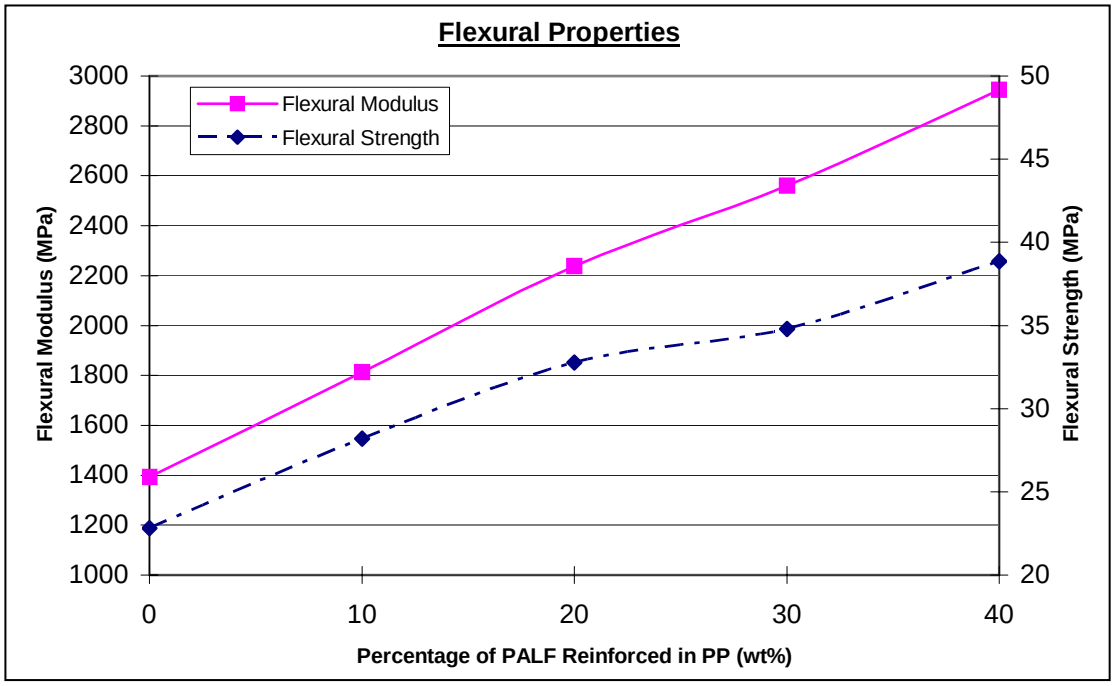


Figure 4.4: Flexural Modulus and Flexural Strength of Short PALF Reinforced PP with Various Fibre Composition

Table 4.2: Variations of Tensile Strength and Flexural Strength with Fibre Loadings in Short PALF-PP

Fibre Content (%wt)	Tensile Strength (MPa)	Flexural Strength (MPa)
0	25.0	22.8
10	16.2	28.2
20	8.6	32.8
30	17.1	34.8
40	2.4	38.9

Table 4.3 presents the flexural properties of long and short PALF-PP with various fibre loadings. Here, same trend of higher flexural strength is shown in long fibre composite compare to short fibre composite. However the flexural modulus of long fibre shows an optimum point in the 10 wt % fibre loading. This may be

attributed to the low interaction and poor dispersion of the fibre in matrix as stated in the works of Asri and Abdul Khalil (2002) and Arib (2003).

Table 4.3: Flexural Properties of Long and Short PALF Reinforced PP with Various Fibre Composition

	Long PALF-PP*		**Short PALF-PP**	
Fibre Content (wt%)	Flexural Strength (MPa)	Flexural Modulus (MPa)	Flexural Strength (MPa)	Flexural Modulus (MPa)
0	66.1	1898	22.8	1392
10	69.5	2004	28.2	1813
20	67.2	1885	32.8	2238
30	68.0	1941	34.8	2561
40	65.6	1849	38.9	2945

* These results obtained from Arib (2003).

4.5 Impact Properties

Figure 4.5 shows the trend of impact strength with different fibre loadings. The impact strength has risen from 3.4 kJ m^{-2} to 9.7 kJ m^{-2} that is with increment of 186 %. It is generally accepted that the toughness of a fibre composite is mainly dependent on the fibre stress-strain behavior especially the strong fibres such as PALF with high failure strain can actually impart high work to fracture on the composites. This is because short fibre composites containing varying volume fractions of strong cellulossic microfibres of different lengths (Pavithran *et al.*, 1987). Nevertheless, this seems to contradict with tensile properties especially tensile strength and elongation at break. The only reason to explain this is that the composite can withstand fast impact load but if tensile stress that is applied slowly, the fibre tends to slip from the matrix and leaving weak points or stress concentrated area. No doubt, these will reduce the elongation at break and give low toughness. Although the impact strength was improving, there was a slight drop at the 10 wt % of short PALF loading (3.1 kJ m^{-2}). Again for this case, the introduction of fibre into

the PP acted as flaw where stresses were easily concentrated thus low energy was enough to initiate cracks and in consequently the composite failed. Devi *et al.* (1997) has reported that the energy-absorbing mechanism of fracture built in the composites includes utilization of energy required to de-bond the fibres and pull them completely out of the matrix using a weak interface between fibre and matrix. In practical interest, a significant part of energy absorption during impact takes place through the fibre pullout process.

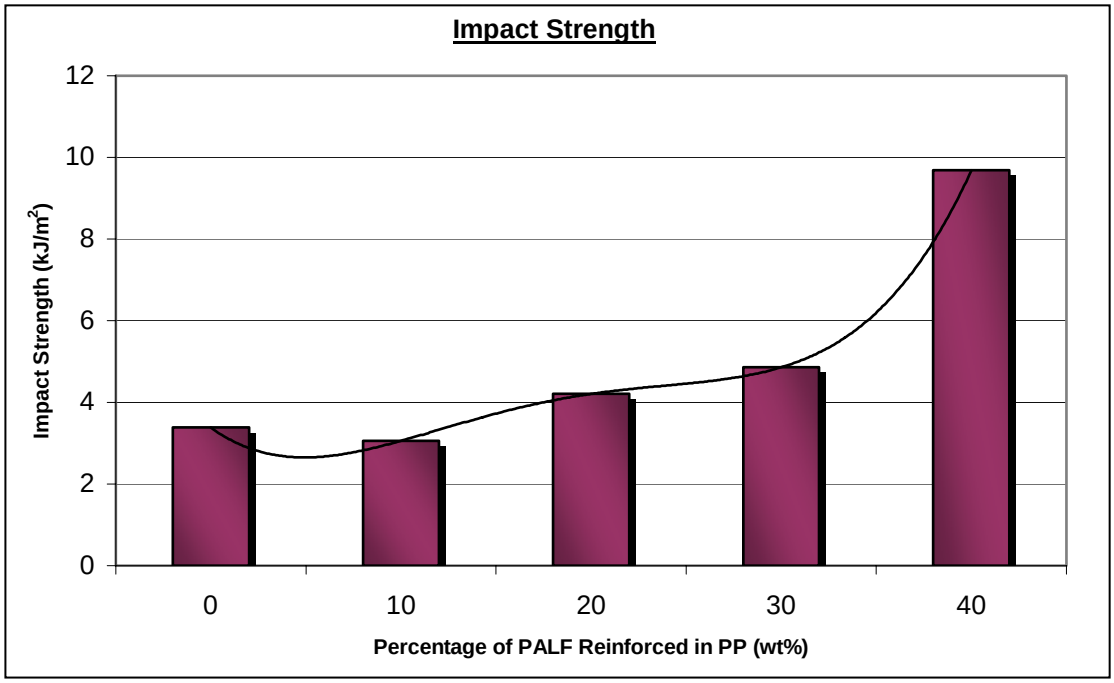


Figure 4.5: Graph of Impact Strength for Varies Short PALF Composition in PP

4.6 Morphological Analysis

Scanning electron micrographs are shown in Figure 4.6 - 4.10 with magnification of 500. In pure PP, the micrograph was seen only with uniform matrix. This served as the control speciment. For 10 wt% treated PALF-PP, there was interference of fibre which tried to hold up the uniform matrix by carrying loads. There was still clear cut of matrix-fibre interface which tends to weaken the adhesion bondings and ease of fibre pull out. These pull outs of fibre can be noticed with the

holes left in the matrix surface. As for 20 wt% treated PALF-PP, the fibre distribution in matrix was not good either and fibre agglomeration as bunch of fibres can be observed. However, the best morphology structure was observed in 30 wt% treated PALF-PP. Fibres are well aligned with the matrix where homogenous stage can be observed. This structure has fewer voids introduced by fibre pull out. In the micrograph, it is hard to notice this. The 40 wt% treated PALF-PP exhibited two regions that are the bulk fibre phase and matrix phase. This clarified that fibre-matrix miscibility and adhesion are weak thus resulting lowering in Young's modulus, tensile strength and elongation at break as discussed earlier.

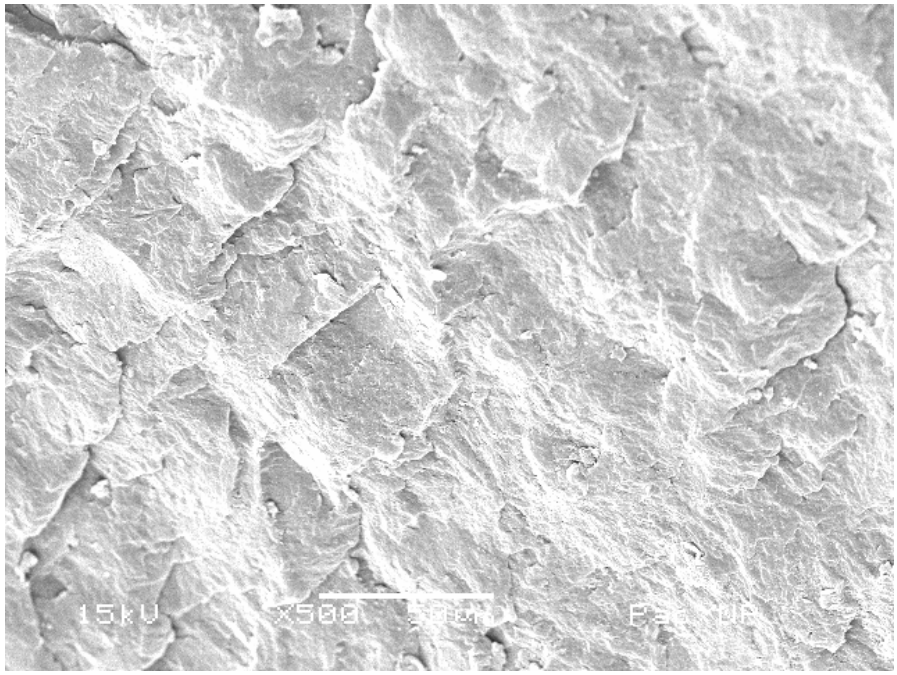


Figure 4.6: Micrograph of Pure PP (0 wt% PALF)

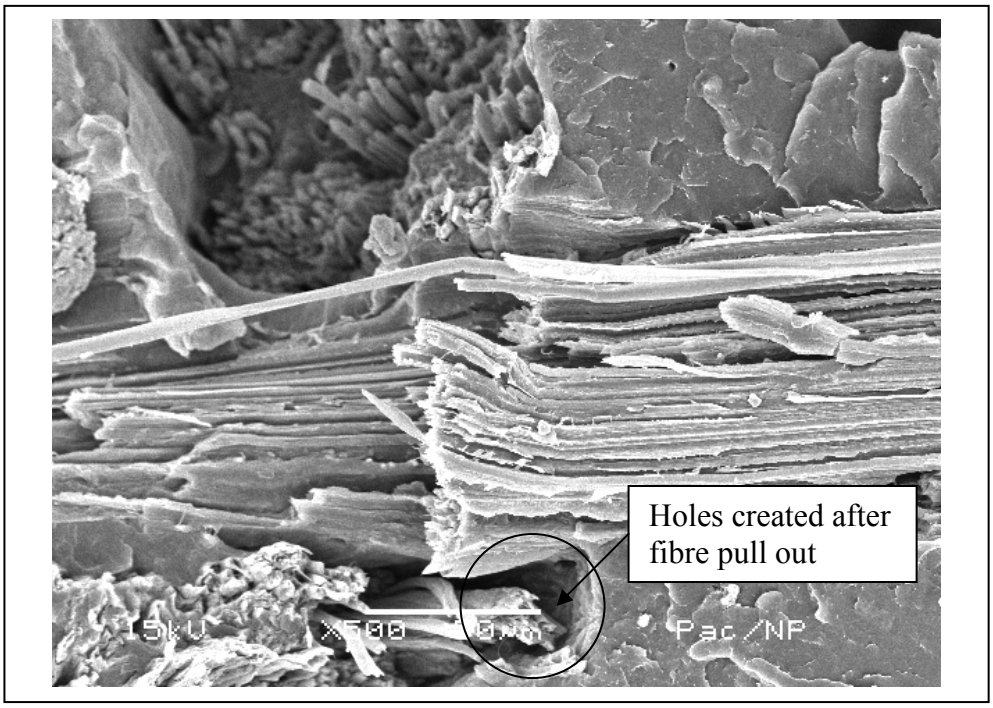


Figure 4.7: Micrograph of treated PALF-PP Composite (10 wt% PALF)

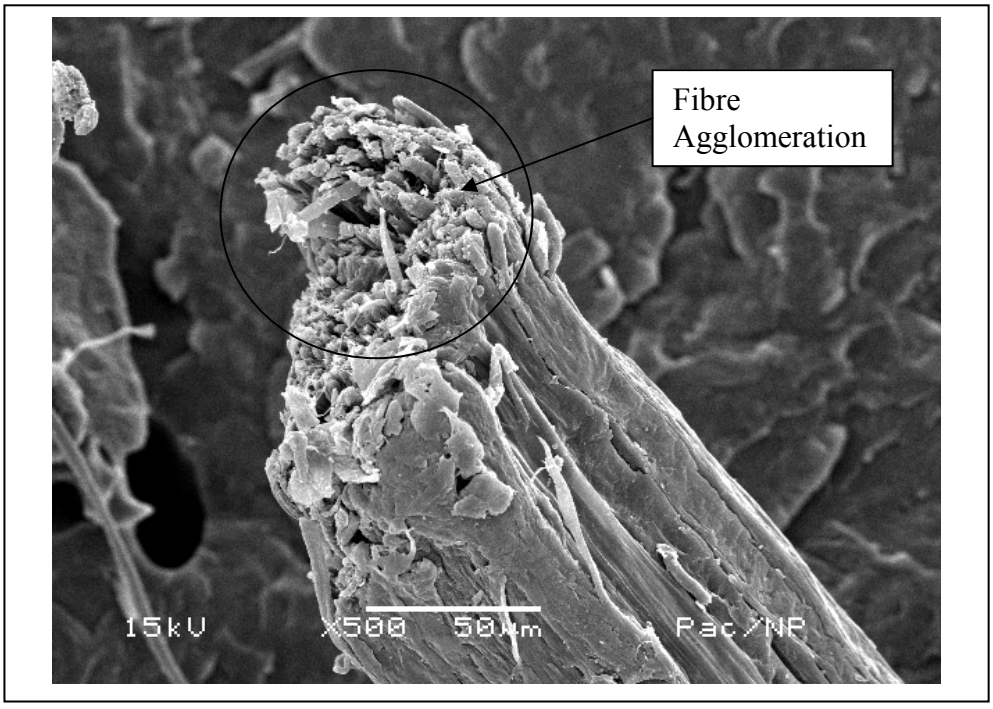


Figure 4.8: Micrograph of treated PALF-PP Composite (20 wt% PALF)

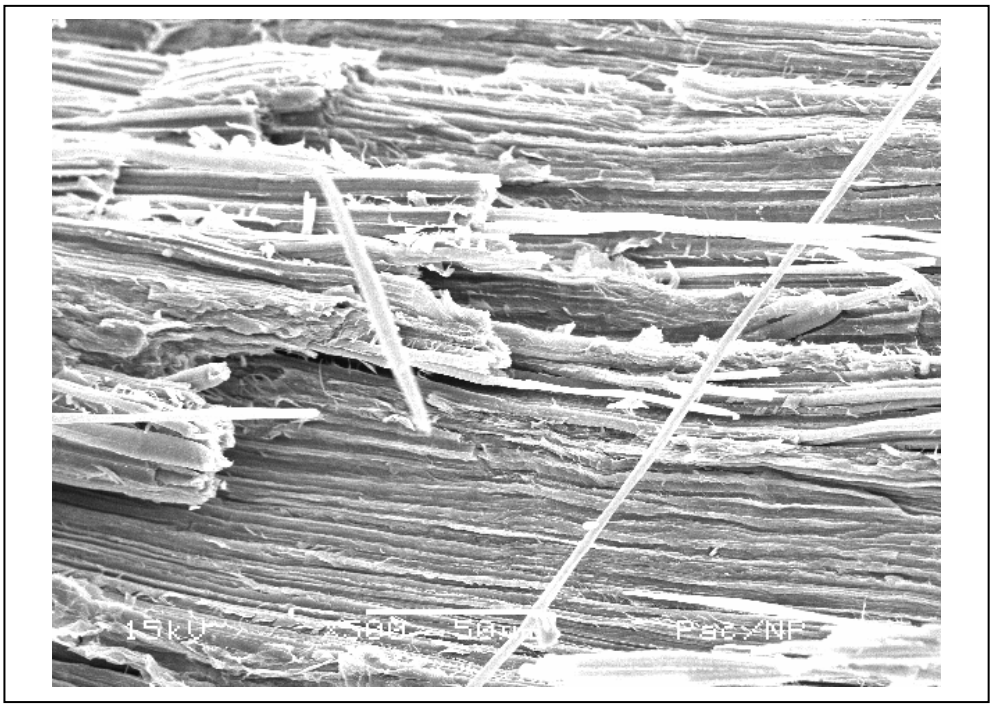


Figure 4.9: Micrograph of treated PALF-PP Composite (30 wt% PALF)

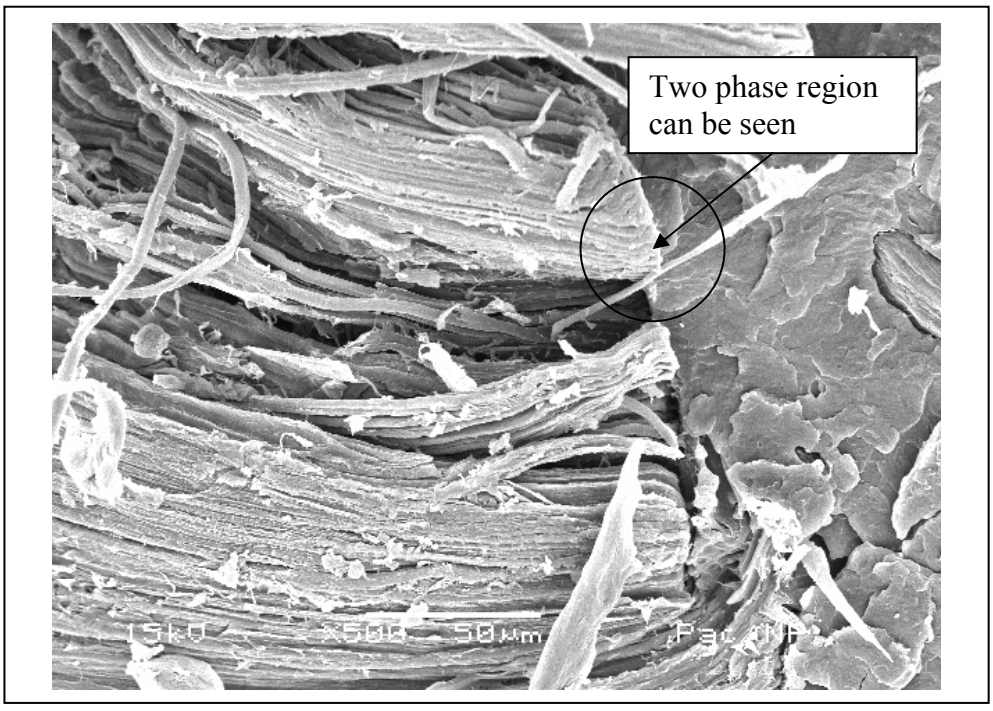


Figure 4.10: Micrograph of treated PALF-PP Composite (40 wt% PALF)

CHAPTER 5

CONCLUSIONS AND FUTURE WORKS

5.1 Conclusions

The results of this present study showed that a useful composite with good properties could be successfully developed using treated PALF as reinforcing agent for the PP matrix. From this, several conclusions can be drawn regarding to mechanical properties of composite to the effect of fibre loadings, namely tensile, flexural and impact properties.

As the PALF loading in PP increased in term of wt%, the Young's modulus increased slowly till 20 wt% and drastically to peak level at 30 wt%. The modulus has increased from 649 MPa to 997 MPa that is about 53% of increment. Further addition of fibre in PP matrix to 40 wt% has reduced the modulus to the initial value due to the fibre incorporated has acted as flaws and this initialized crazing and created stress concentration area. Conversely, it is found that the tensile strength declined as the fibre concentration in composite increased. The increase of fibre-to-fibre interaction and dispersion problem in matrix has contributed to this phenomenon. In a similar vein, there is a decrement observed for elongation at break for higher PALF loadings. The addition of stiff fibre has interrupted the PP matrix and uneven distribution made the composite more brittle instead of ductile.

The flexural modulus and flexural strength were increasing gradually as the PALF content in composite increased. It is observed that the flexural strength increased from 22.8 MPa to 38.9 MPa and flexural modulus increased from 1392 MPa to 2945 MPa respectively for pure PP to 40 wt% fibre. This gradually increase trend has shown that PALF can withstand bending forces to a great extent since their high crystalline fibrils content are strong and can share the load applied in matrix effectively.

Identical to flexural properties, impact properties also indicates a rise from 3.4 kJ m^{-2} to 9.7 kJ m^{-2} as the fibre content increased. This is because this natural fibre is considered oriented short fibre composites with strong cellulossic microfibres of different lengths (Pavithran *et al.*, 1987). However the impact strength experienced a slight drop at the initial incorporation of 10 wt % short PALF loading (3.1 kJ m^{-2}). One interpretation of this is the low content of PALF in PP has acted as flaw and can easily initiate cracks in this stress concentrated area, especially the fibre end.

Finally to summarize everything, treated PALF has enhanced tensile properties in Young's modulus, flexural as well as impact properties of the PP. The study has demonstrated the optimum fibre loading for peak performance is at 30 wt%. Fibre matrix interaction is well adhered and compatible with the use of coupling agent at this concentration of fibre. Splitting, peeling and pull out of fibre is not obvious in the SEM micrographs for the 30 wt% but rather a more corrugated fibre. Synergistic results can be obtained through incorporation of PALF in PP matrix compared to single component of PP or PALF.

5.2 Recommendations and Future Works

This study may be more applicable and better if the following suggestions are done.

(i) Chemical treatment of fibre can be done with isocyanate like PMPPIC. Better performance of PALF-PP composite is expected as this is the best chemical treatment suggested by previous research works.

(ii) Solution mixing is suggested to replace melt mixing in composite preparation since PALF will be well adhered in PP matrix, thus enhancing the mechanical properties.

(iii) Compounding of short fibre and PP should be done in twin screw extrusion to give better mixing effect as it provides greater control of mixing, shear and conveying properties by regulating the amount of clearance between the screws.

(iv) Currently the production of laminate composite via hot press is not accurate enough. Suitable and well design mould should be prepared for this. However it is recommended that the use of injection moulding to prepare testing sample is more precise as it reduced much of human factor's error, such as machining the composite to testing specimen which has critical dimensions.

(v) Actually in pendulum impact test, there is another one known as Charpy Impact Test. Charpy method is a better option because the specimen does not have to be clamped and, therefore, it is free of variations in clamping pressures.

The results of this study suggested a number of new avenues for research in future. They are:

(i) Determination of chemical constituents inside the local abundant pineapple leaf fibre to the extent of chemical content and its effects to certain properties.

(ii) The work should be extended to study other properties such as creep, fatigue, shear strength, chemical resistance and electrical properties.

(iii) The usage of different types of chemical treatment and coupling agents can be studied for PALF-PP composite.

(iv) Besides polyester, LDPE, HDPE and PP, other comoditive polymeric matrix system can be studied. Perhaps PS which is brittle in nature can have great synergistic effect after incooperation with PALF.

(v) Hybrid composite comprising other fibre (such as fibre glass) besides PALF can be studied as this will definitely yield better performance of composite system.

(vi) Attention to this lignocellulosic materials also can be extended to the biodegradibility aspect. Contolled biodegradability after effectice use is another important factor in favour of biofibre composite.

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