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PROCESSING AND MECHANICAL PROPERTY TESTING OF PAPER FIBER AND HIGH DENSITY POLYETHYLENE COMPOSITES

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ABSTRACT

PROCESSING AND MECHANICAL PROPERTY TESTING OF PAPER FIBER AND HIGH DENSITY POLYETHYLENE COMPOSITES

By

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The utilization of waste paper can be increased by combining paper fibers with thermoplastics like High Density Polyethylene (HDPE) into a composite material. Two kinds of paper fibers, mixed and deinked paper fibers, were used as fillers in composites. The mechanical and physical properties were studied by varying the fiber content and using different kinds of fibers at the same processing conditions. HDPE and paper fibers were combined in a twin-screw extruder to form the composite, and then compression molded. Tensile properties, Izod impact strength and water absorption were evaluated following ASTM standard procedures. The dispersion of fibers was seen by scanning electron microscope (SEM). The results show that the addition of paper fibers to HDPE causes a decrease in tensile strength and impact strength, but an increase in tensile modulus. Water was more absorbed due to the addition of paper fibers. SEM showed that the fibers did not disperse in the matrix very well.

Mixed paper fiber composites gave better results in strength when compared with deinked paper fiber composites. Composite strength was lower than aspen wood fiber composites from prior experiments, but water absorption was lessened.

To my parents , Pattama and Chuan Chotipatoomwan

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INTRODUCTION

Nowadays, paper products and packages are in high use in industrial, commercial, and residential applications. Consequently, these cause us a lot of trouble in decreasing the amount of fiber resources used because wood fibers and pulps are the main sources for paper manufacturing. Moreover, controlling the high quantity of paper wastes is also needed to reduce the environmental problems, such as the concern over the volume of solid waste being directed to landfill sites.

In the United States, all types of solid wastes amount to about 60,415 pounds of waste per person per year (Selke, 1994). These solid wastes are classified as water pollutants, industrial waste, wastewater treatment, air pollutants, oil spills, mining, demolition waste, agricultural waste, hazardous waste, and municipal solid waste (MSW).

Packaging wastes are one part of MSW, about 1/3 of the total, and paper and paperboard are the highest percentage of this part. Paper products have traditionally been disposed to landfills, and this is becoming increasingly expensive with landfill space becoming less available. The recycling of recovered paper, not a new phenomenon, could be the way to solve these problems. There are two measures about which we are concerned. Utilization and recovery rates are the factors which tell about the materials effect on the environment. The higher percentage they are, the less problem it is.

First of all, the utilization rate is measured from the amount of wastepaper used in the production of a specific product, or product sector, or by the industry.

$$\text{Utilization rate} = \frac{\text{Waste paper used (tons)} \times 100}{\text{Paper or board production (tons)}} \qquad (1)$$

For example, the consumption of paper and board from around the world in 1992 was 245.6 million tons, but the amount of wastepaper consumption was only 96 million tons (Mckinney, 1995). These means that the utilization rate is about 31%.

Another factor is the recovery rate, which is a measure of recovery of wastepaper , and is given as a proportion of the total paper and board consumption.

$$\text{Recovery rate} = \frac{\text{Wastepaper recovered (tons)} \times 100}{\text{Apparent consumption of paper and board (tons)}} \qquad (2)$$

Because of the high volume of solid waste and the lack of adequate fiber sources, many countries have introduced programs, legislation and regulations to promote waste fiber recycling. Therefore, the paper recovery rate has increased each year.

Two primary indices, recovery and utilization rates, are used to compare the level of recycling in various countries. The difference between these indices is that the utilization rate is the comparison of the amount of secondary fiber used in paper/board production with the total fiber used, whereas recovery rate is the comparison of wastepaper recovered for reuse with paper consumed (Smook, 1992). These two indices do not need to be equivalent, and depend on major factors, such as the amount of forest resources, and the paper industry in each country. For example, Sweden is the leading paper exporting country, so the utilization rate is understandably less (Smook, 1992).

Table 1. Recovered Paper (Thousand tons)

Country	1989	1994
Canada	1652	2458
Finland	684	1472
France	2877	3514
Germany	5663	9758
Italy	1452	2278
Japan	n/a	14908
Norway	157	297
Portugal	291	3
Spain	1591	1816
Sweden	890	990
Switzerland	n/a	754
Turkey	n/a	425
United Kingdom	2880	4048
United States	24664	35100

Source : OECD 1997

Table 2. Apparent Recovery and Utilization Rates (1992)

Country	Utilization (%)	Recovery (%)
USA	33.1	38.7
Germany	52.1	50.6
Japan	52.7	51.1
The Netherlands	70.5	54.7
Sweden	14.3	49.7
Australia	45.8	36.8
South Korea	69.6	43.2
UK	60.2	33.9
Denmark	96.6	37.6

Source : McKinney 1995

In Japan, the wastepaper recovery rate from 1953 to 1991, increased from 19.6% to 50.8% (McKinney, 1995) because of the lack of forest resources. The United States has a lower recovery rate. In 1994, the recovery rate was just 40% (OECD, 1997). However, this is slightly above the world average.

Even if the utilization rate is high, it does not mean that paper fiber can be processed many times in a recycling system, because significant losses of both fiber substance and strength occur during each recycling. It is considered that a fiber can be recycled only four times before the loss in some quality, such as fiber substance and strength (Smook, 1992). The effect of multiple recycling operations on fiber characteristics had been studied from the late 1960's to the late 1970's. Smook summarized results from one study on the effect of repeated recycling of newsprint on individual fiber strength and on the bonding strength between fibers. Both strength types were decreased, and the bonding between fibers was dramatically lost. With each drying and slushing cycle, the fibers become less flexible and less permeable to water, and therefore do not conform as well as virgin fibers. Cumulative loss of hemicellulose from the fiber surfaces also contributes toward reduced bonding.

Besides ordinary waste paper, waste polymer-coated paper is a concern, especially paper coated with LDPE which is used to produce such products as aseptic packages, paper milk cartons, frozen food boxes, and paper plates and cups.

In 1992, researchers at the USDA Forest Service Forest Products Laboratory (FPL) were aware of the problems associated with LDPE. Then, they considered the ways to reuse these materials. Finally, there were three ways which were proposed: (a) burning it for fuel, (b) drying and separating the paper fiber from the LDPE so that each material could be recycled using existing technologies, and (c) making a fiber/plastic composite of the material (English and Schneider, 1994)

However, the first and second options were not chosen because of environmental concerns and the difficulty in separation. The FPL researchers decided making a paper fiber/plastic composite was the most attractive option.

Generally, in composite materials, wood and paper fiber have been used as a reinforcing fiber in thermosetting plastics. On the other hand, thermoplastics, like LDPE, have recently been mixed with wood and paper fiber also. Moreover, several billion pounds of fillers and reinforcements are used annually in the plastic industry (Katz and Milewski, 1987).

There are many reasons that a fiber/plastic composite was produced.

1) Material cost saving : As plastic materials have relatively high cost, the fibers which have low cost can help by reducing the quantity of plastic used.
2) Improving some physical properties of thermoplastics : The addition of wood or paper to thermoplastics increases stiffness and strength and reduces creep.
3) This material can be used in automotive, building, appliance, and other applications.

However, there are still some drawbacks occurring from this material also. For example:

1) Possibility of degradation of fibers : fibers can lose their properties due to high temperatures in processing.
2) Limited type of thermoplastics: just some kinds of thermoplastics can be used with agro-fibers, for example polyethylene (PE), polypropylene (PP), polyvinyl chloride (PVC), and polystyrene (PS), because it is difficult to mix fibers with thermoplastic due to their high melting points and high solution viscosities.
3) Moisture absorption: by nature, fibers can absorb moisture, and this can result in swelling of the fibers. However, the absorption of moisture by the fibers is minimized in the composite due to encapsulation by the polymer.

The primary objective of this research was to utilize the wastepaper in the form of paper fibers in the process of composite materials, resulting in manufacturing cost savings, decreasing of environmental problems, and improvement of properties in the composites.

As the cost of HDPE resin is higher than that of the fiber, and the quantity of HDPE used in the composite decreases as the fiber ratio increases, the cost can be cut down in this way. Moreover, the amount of wastepaper can be reduced by using it as a raw material in this kind of composite.

However, some properties are changed by adding paper fibers into the HDPE matrix. In this research, tensile strength, yield strength, % elongation, modulus of elasticity, and Izod impact strength were tested by comparing the effects on the composite of two kinds of paper fibers, high-grade deinked and mixed paper fibers.

Moisture absorption and dispersion of fibers in the matrix are important to know. Even though the fibers are encapsulated by the polymer matrix, they can swell up by absorbing moisture outside. ASTM 570-81 determines the amount of moisture (in percentages) that the materials absorb as a function of time. To see how fibers disperse in the composites, scanning electron microscopy (SEM) can be used to show how fibers are mixed into the polymer matrix.

Lastly, data on aspen wood fibers from the earlier experiments by JoAnna Denise Childress (1991), were compared with the results from paper fibers, as they both functioned as the reinforcement in composites. The comparison between the fibers was made at the ratio of 40% by weight in HDPE.

CHAPTER 1

LITERATURE REVIEW

1) Background on composite materials

Generally, a structural composite consists of three phases. One of the phases is the reinforcement, which is usually discontinuous, stiffer, and stronger. The continuous phase, which is less stiff and weaker, is called the matrix. Finally, the interface is the chemical or other interactions between the reinforcement and the matrix.

Each phase has different roles that depend on the application and type of the composite materials. In this project, the chosen reinforcement, paper fibers, is usually in the form of short fibers, and it provides stiffness, whereas the matrix is the main load-bearing constituent governing the mechanical properties of the material. Additionally, the matrix can protect the fiber from abrasion and the effects of the environment. The interface can play an important role in controlling the failure mechanisms, fracture toughness, and overall stress-strain behavior of the material.

2) Reinforcement

Paper fibers made from waste papers are used as the reinforcement in these experiments. Waste papers have been divided into five grades by the American Forest & Paper Association (OECD, 1997).

a) Mixed paper : Mixed papers, super mixed papers, office papers (if not deinked or of suitable quality to be used as a pulp substitute), telephone directories, magazines and catalogues, recycled boxboard cuttings, tissue paper converting scrap if predominately composed of recycled fiber, mill wrappers, specialty grades and all other grades not elsewhere specified.

b) Newspapers : Old newspapers (ONP), special news (including deinked quality) overissue news, white blank news, ground-wood computer printout publication blanks, mixed ground-wood and flyleaf shavings, coated ground-wood sections.

c) Corrugated : Old containers both corrugated and solid fiber, container plant cuttings, kraft paper and bags, kraft bag clippings, carrier stock and carrier stock clippings.

d) Pulp substitutes : Unprinted paper and board that has not been coated or adulterated in any way; included in this category are tabulating cards, white and semi-bleached sheets, cuttings, shavings, or trimmings.

e) High-grade deinked : Bleached chemical grade office papers and computer printout to be deinked, bleached sulphite and sulphate cuttings including tissue paper converting scrap if predominantly composed of bleached chemical pulp fiber, coated book stock. Printed grades, if deinked, are reported as high grade deinking.

In this case, fibers from mixed paper and from high grade deinked paper were compared in mechanical properties when they were combined in composites. Moreover, the effects of fiber content, fiber length, dispersion, and ability of water absorption were investigated.

Reinforcing fibers are classified into three categories, which are particulate, continuous, and discontinuous fibers (Daniel and Ishai, 1994).

Particulate composites: They consist of particles of various sizes and shapes randomly dispersed within the matrix. Because of the randomness of particle distribution, these composites can be regarded as quasi-homogeneous on a scale larger than the particle size and spacing and quasi-isotropic. Particulate composites may consist of either nonmetallic or metallic particles in nonmetallic or metallic matrix. Examples include concrete and glass reinforced with mica flakes.

Discontinuous or short fiber composites: They contain short fibers or whiskers as the reinforcing phase. These short fibers, which can be fairly long compared with the diameter, can be either all oriented along one direction or randomly oriented. In the first instance, the composite material tends to be markedly anisotropic, or orthotropic, but in the second, it can be regarded as quasi-isotropic.

Continuous fiber composites: They are reinforced by long continuous fibers and are the most efficient from the point of view of stiffness and strength. The continuous fibers can be all parallel, can be oriented at right angles to each other, or can be oriented along several directions.

As paper fibers are a kind of short fibers, they are classified as discontinuous fibers. Addition of short fiber reinforcement to thermoplastic materials can be used to enhance physical properties and performance characteristics. There are many factors that influence the properties of composites, such as fiber type, length of fiber, length to diameter ratio, fiber alignment, interface, matrix resin morphology, processing procedures and environmental effects.

3) Polymer matrix

A polymer is defined as a long-chain molecule containing one or more repeating units of atoms, joined together by strong covalent bonds (Mallick, 1988). In general, they are divided into two broad categories, thermoplastics and thermosets. These two kinds of matrices have both advantages and disadvantages, when they are reinforced into composite materials.

Thermoset polymers in composites normally give thermal stability and chemical resistance. They also exhibit much less creep and stress relaxation than thermoplastic polymers. Then, thermosetting materials are particularly suitable as composite materials due to their relative ease of fabrication and good adhesion characteristics. However, their disadvantages are their limited storage life (before the final shape is molded) at room temperature, long fabrication time in the mold, and low strains to failure which also cause them to have low impact strength.

On the other hand, thermoplastic polymers in composites will give high impact strength and fracture resistance, which in turn impart excellent damage tolerance characteristics to the composite materials. Moreover, thermoplastic polymers have higher strains to failure, so they provide a better resistance to matrix microcracking in composite laminates. Even though thermoplastic polymers have a lot of advantages, they still have been developed slowly compared to thermoset matrices (Mallick, 1988). Because of their high melt or solution viscosities, incorporation of continuous fibers into thermoplastics is difficult.

From several experiments, it was determined that some polymers are susceptible to reinforcement, others are not. Especially, it is difficult to incorporate continuous fibers into most thermoplastics, such as polycarbonate and nylon, due to their high melt and solution viscosities (Mallick, 1988). This means the requirements for filler particle size depend upon the polymer matrix type.

4) Interface

Berlin et al (1986) studied the effect of adhesion at the interface in composites, which is divided into two categories. First, in the case of good adhesion between the matrix and fibers, the maximum stress that can be transmitted from the matrix to the fiber is equal to the shear yield point of the matrix, τ_m, for a plastic matrix, and equal to the shear strength of the matrix for a brittle material. Secondly, in the case of poor adhesion, the maximum stress transmissible from the matrix to the fiber will be smaller than τ_m and equal to the strength of adhesion (Berlin et al, 1986).

In the complete absence of adhesion, a very small stress applied to the matrix would cause detachment of the matrix material from the fiber surface with the formation of voids. No stresses at all would be transmitted to the fiber in this case.

The mechanism of stress transfer at the interface in composites is much more complex. It is important to distinguish between normal stress and shear stress. Shear stresses will transmit tensile stress to the fibers along the fiber orientation. Normal stresses arise on the side surfaces of the fibers due to the residual thermal stresses in the composite and the different Poisson's ratios (ν) of the matrix and fiber.

In continuous fiber-reinforced composites, the resistance to shear between the matrix and fiber has a relatively small effect on the composite strength under tension along the fibers. However, it does have a certain effect at stresses close to the ultimate stress, when the weaker fibers begin to break and stress transmission through the matrix begins to play an important role. When tension is applied across the fibers, it is not the resistance to shear but the resistance to separation (σ_{mf}) which begins to play the major role.

In general, at high ratios of fibers in composites, the strength of reinforced composites will depend on σ_{mf}, the maximum values of tensile stress in the composites (σ_c) will decrease because a matrix can come off the transversely oriented fibers and the resultant voids will initiate crack development and, therefore, the composites will not have a high mechanical strength.

Berlin et al (1986) classified three factors that needed to be considered: void size, void content, and the stress causing dewetting, which causes the fibers to not adhere to the matrix. The void size is determined by the filler particle size. The smaller the particles, the smaller will be the voids resulting from dewetting of the matrix. The void content is determined by the volume fraction of the filler. Finally, the stress required to cause dewetting depends on the adhesion strength between the filler particles and the matrix.

5) Prediction of properties

Theoretically, calculations for tensile strength, modulus, and other properties of fiber-reinforced composites are based on the fiber volume fraction in the material, which can be determined from Equation 3 (Berlin et al, 1986).

$$V_f = \frac{W_f/\rho_f}{(W_f/\rho_f) + (1-W_f)/\rho_m} \qquad (3)$$

where: W_f = fiber weight fraction

$(1-W_f)$ = matrix weight fraction

ρ_f = fiber density

ρ_m = matrix density

The composite density (ρ_c) is another factor related to the fiber weight fraction (W_f). It can be calculated by the following equation.

$$\rho_c = \frac{1}{(W_f/\rho_f) + (1-W_f)/\rho_m} \tag{4}$$

During the processing of composites, air or other volatiles may be trapped in the material. This can cause voids, which will affect the mechanical properties of the composite materials. Void content can be estimated by Equation 5.

$$V_v = \frac{\rho_c - \rho}{\rho_c} \tag{5}$$

where: V_v = volume fraction of voids

ρ_c = theoretical density (as calculated in equation 4)

ρ = actual density, measured experimentally on composite specimen

A high void content (% by volume) usually leads to lower fatigue resistance, greater susceptibility to water diffusion, and increased variation in mechanical properties.

In the case of anisotropic materials under uniaxial tension along a direction, the axial and transverse deformation (strain) are given by Equations 6 and 7 respectively (Berlin et al, 1986).

$$\epsilon_x = \frac{\sigma_x}{E_x} \tag{6}$$

$$\epsilon_y = \frac{-\nu_{xy}\sigma_x}{E_x} \tag{7}$$

where: σ_x = axial normal stress

E_x = axial modulus in x-direction

ν_{xy} = Poissons ratio associated with loading in the x-direction and strain in the y-direction

Additionally, Poisson's ratio (ν_{xy}) can be estimated by Equation 8

$$\nu_{xy} = \frac{\eta_{xs}\sigma_x}{E_x} \tag{8}$$

where: η_{xy} = shear coupling coefficient (the first subscript denotes normal loading in the x-direction; the second subscript denotes shear strain)

Normally, fiber-reinforced composites are microscopically inhomogeneous and orthotropic (Mallick, 1988). Thus, the mechanics of fiber-reinforced composites are far more complex than those of conventional materials. Predictions of mechanics of fiber-reinforced materials are divided into two different approaches.

1) The micromechanics approach: Equations describing the elastic and thermal characteristics of a lamina, which is the incorporation of a large number of fibers into a thin layer of matrix, are based on micromechanics formulations.
2) The macromechanics approach: The studied material is assumed to be homogeneous. Equations of orthotropic elasticity are used to calculate stresses, strains, and deflections.

Mallick (1988) considered the mechanics of fiber-matrix interactions in a unidirectional lamina. Then, he proposed the basic assumption, in which fibers are distributed throughout the matrix and bonded to the matrix perfectly, and voids cannot occur. Applied loads are used in either the longitudinal or transverse direction. As continuous parallel fibers were used to study longitudinal tensile loading, bonding between the fibers and matrix is assumed to be perfect.

Then, $\quad \epsilon_f = \epsilon_m = \epsilon_c \quad$ (9)

where ϵ_f, ϵ_m, ϵ_c are the longitudinal strains in the fibers, matrix, and composite respectively. Since both fibers and matrix are elastic, the stresses can be shown to be the following (Mallick, 1988):

$$\sigma_f = E_f \epsilon_f = E_f \epsilon_c \quad (10)$$

$$\sigma_m = E_m \epsilon_m = E_m \epsilon_c \quad (11)$$

When a tensile force is applied on a composite material, it will be shared by the fiber and matrix. The total force (P_c) can be written as:

$$P_c = P_f + P_m \quad (12)$$

Since the force or load is equal to stress (σ) × area (A), so equation 12 can be rewritten as

$$\sigma_c A_c = \sigma_f A_f + \sigma_m A_m \quad (13)$$

or

$$\sigma_c = \frac{\sigma_f A_f}{A_c} + \frac{\sigma_m A_m}{A_c} \quad (14)$$

where σ_c = average tensile stress in the composite

A_f = net cross-sectional area for the fibers

A_m = net cross-sectional area for the matrix

The total area of the composite will be the area of the fibers plus matrix:

$$A_c = A_f + A_m \quad (15)$$

Since $V_f = A_f/A_c$ and $V_m = A_m/A_c$, from equation 14

$$\sigma_c = \sigma_f V_f + \sigma_m V_m$$

$$= \sigma_f V_f + \sigma_m(1-V_f) \quad (16)$$

Since modulus of elasticity equals the ratio of stress and strain,

$$E = \sigma/\in \quad (17)$$

then, the longitudinal modulus for the composite can be written as:

$$\sigma_c = E_f V_f \in_f + E_m V_m \in_m \quad (18)$$

Equation 18 can be rewritten in the form of the rule of mixtures as equation 19.

$$E_c = E_f V_f + E_m(1-V_f) \quad (19)$$

Birley et al (1992) said the rule of mixtures gave an upper-bound limit for stiffness, which is related directly to the volume fraction (V_f) of the reinforcement phase. Fiber content is calculated on a weight fraction basis with loading up to 40% (by weight) being typical. In general, the longitudinal modulus is actually less than that predicted by equation 19.

Wherever stresses are applied at an angle to the fiber-axis, the modulus diminishes to a value between the limits defined by equation 19, and an equivalent expression for the transverse modulus.

$$E_t = \frac{1}{(V_f/E_f) + (V_m/E_m)} \quad (20)$$

Calculating from equation 20 is usually inaccurate and pessimistic. Then, it can be shown that a semi-empirical modification of equation 20 yields (Birley, et al, 1992).

$$\frac{1}{E_t} = \frac{1}{V_f + \eta_t V_m} [\, V_f/E_f + \eta_t V_m/E_m] \quad (21)$$

where η_t = the stress partition parameter, for which experimental data has shown 0.5 to be an accurate representation.

In general, the fiber failure strain is lower than the matrix failure strain. Mallick (1988) assumed that all fibers had the same length, and the tensile rupture of fibers precipitated the tensile rupture in the composites, Thus, using equation 16, the longitudinal tensile strength (σ_{Ltu}) of a unidirectional continuous composite can be estimated as:

$$\sigma_{Ltu} = \sigma_{fu} V_f + \sigma_m'(1-V_f) \qquad (22)$$

where σ_{fu} = fiber tensile strength (assuming a single value for all fibers)

σ_m' = matrix stress at the fiber failure strain (at $\epsilon_m = \epsilon_{fu}$)

In the case of discontinuous parallel fibers, tensile load is transferred to these fibers by a shearing mechanism between the fibers and matrix. Since the matrix has a lower modulus, the longitudinal strain in the matrix is higher than that in adjacent fibers.

If a perfect bond is assumed between the two constituents, the difference in longitudinal strains creates a shear stress distribution. The normal stress distribution in a discontinuous fiber is calculated by the following equation.

$$\frac{d\sigma_f}{dx} = \frac{4\tau}{d_f} \qquad (23)$$

where σ_f = longitudinal stress in fiber at a distance x from one of its ends

τ = shear stress at the fiber/matrix interface

d_f = fiber diameter

To determine the normal stress distribution in the fiber at either one of its ends, equation 23 can be derived into the following equation (Mallick 1988):

$$\sigma_f = \frac{4}{d_f}\int \tau dx \qquad (24)$$

Normally, the interfacial shear stress is assumed to be constant. Then,

$$\sigma_f = \frac{4\tau_i x}{d_f} \tag{25}$$

where τ_i = interfacial shear stress

A composite lamina contains discontinuous fibers, the fiber stress is not uniform. It is zero at the ends and builds up linearly to the maximum value at the central portion of the fiber. Then, the maximum fiber stress can be estimated by equation 26.

$$(\sigma_f)_{max} = \frac{2\tau_i L_t}{d_f} \tag{26}$$

where $x = L_t/2$ = load transfer length at each fiber end. For a given fiber diameter and fiber/matrix interfacial condition, a critical fiber length (L_c) is calculated from equation 26 as

$$L_c = \frac{\sigma_{fu} d_f}{2\tau_i} \tag{27}$$

where σ_{fu} = ultimate fiber strength

L_c = minimum fiber length required for the fiber stress to be equal to the fiber ultimate strength at its midlength.

This ultimate value is also valid when stress is transferred due to friction between the matrix and fibers. The critical length of fibers can be used to predict the failure of composites by comparing it to the fiber length (L_f).

- When $L_f < L_c$, the maximum fiber stress may never reach the ultimate fiber strength. In this case, either the fiber/matrix interfacial bond or the matrix may fail before fibers achieve their potential strength.

- When $L_f > L_c$, the maximum fiber stress may reach the ultimate fiber strength over much of its length. However, over a distance equal to $L_c/2$ from each end, the fiber remains ineffective.

The critical length can be controlled by increasing or decreasing τ_i. A matrix compatible coupling agent may increase τ_i, which in turn decreases L_c. If L_c can be reduced relative to L_f through paper coupling agents, effective reinforcement can be achieved without changing the fiber length.

To investigate impact behavior, it is assumed that the addition of solid fillers to plastic matrices makes composites more brittle, if the filler content (by volume) is around or above 0.2 (Berlin, et al, 1986).

As impact strength is defined as the capacity to absorb and dissipate energies under impact shock loading (Mallick, 1988), the energy-absorbing mechanisms in the composite included the following conditions:

1) Utilization of the energy required to debond the fibers and pull them completely out of the matrix.
2) Use of a weak interface between the fiber and the matrix.

In most fiber-reinforced plastics, a significant part of the energy absorption during impact takes place through the fiber pullout process. The largest amount of energy involved, and so the greatest toughness, occurs when the length of the fibers is equal to the critical length (L_c). The variation of impact strength with fiber length was calculated by using a model based on condition 1. Fibers shorter than L_c will be pulled out from the matrix rather than broken when a crack passes through the composite. The fracture energy will largely be a combination of the work needed to debond the fibers from the matrix and the work done against friction in pulling the fibers out of the matrix. The fracture energy (U) arising from fiber pullout is given by equation 28 and 30.

For $L < L_c$

$$U_1 = \frac{v\tau L^2}{12d} \tag{28}$$

where d = the fiber diameter

τ = the interfacial friction stress

v = the volume fraction of the fiber

Hence, $U_1 \propto 1/L^2$ for $L < L_c$

when L = Lc, the energy reaches a maximum and will have a value:

$$U_{max} = \frac{v\tau L_c^2}{12d} \tag{29}$$

In case of $L > L_c$, only a portion of the fibers will pull out, and then the energy is given by

$$U_2 = \frac{v\tau L_c^3}{12dL} \tag{30}$$

The impact energy or strength decreases as:

$$U_2 \propto 1/L$$

This decrease in impact strength for composites will happen with the fiber length shorter than the critical length. However, the impact strength of composites is found to increase linearly with the weight fraction of fiber (Devi et al, 1997).

Prior research

There are a lot of studies on the properties of composites. Most fillers in the research reviewed here, were from natural resources like wood fibers, such as jute, sisal, and pineapple leaf, including paper fiber. Several kinds of polymer matrices, thermoplastics and thermosets, were combined into the composites. The fiber, matrix, and the adhesion between them can affect the mechanical properties of composites.

Gogoi (1989) investigated the effects of fiber pre-treatment, screw configuration of a twin-screw extruder, and compounding temperature on the mechanical properties of the composite. Aspen fibers were combined with recycled high density polyethylene, and mixing conditions were varied. The tensile strength decreased gradually with increasing fiber content, whereas the tensile modulus increased with fiber content. The impact strength was decreased compared to that of recycled HDPE, but flexural strength was increased when the fibers were added.

Raj et al (1990) investigated the effect of aging on mechanical properties of linear low density polyethylene (LLDPE)-glass fiber, mica, and wood fiber composites. The effect of aging on mechanical properties of composites was examined under different conditions: (1) exposure at 105 C for 7 days; and (2) immersion in boiling water for 4 hrs. Samples with glass fibers showed the best results with regard to tensile strength, elongation, and fracture energy. LLDPE with mica produced poor results compared to wood fiber composites. Dimensional stability of LLDPE-wood fiber composites, after boiling water treatment, was inferior to mica and glass fiber composites.

Felix and Gatenholm (1991) studied the nature of adhesion in composites of modified cellulose fibers and polypropylene. Cellulose fibers were surface modified with polypropylene-maleic anhydride copolymer. Then, the modified fibers were compounded with polypropylene, and composites with various amount of fibers were manufactured by injection molding. All mechanical properties were improved when treated fibers were used. Scanning electron microscopy (SEM) showed improved dispersion, wetting of fibers, and adhesion. The study found that the surface modifying agent is covalently bonded to the fibers through esterification. The degree of esterification is enhanced by activating the modifying agent before fiber treatment.

Raj et al (1992) studied mechanical properties of polyethylene-organic fiber composites. The selected organic fibers were obtained from blending peanut hulls and pecan shells. Composites were made by using a compression-molding technique. Studies of variations in molding temperature (145-180 °C) and fiber concentration (0-40% by weight) were correlated to the mechanical properties of the composites (tensile strength, elongation, fracture energy, modulus, and impact strength). In untreated nut shell composites, tensile strength decreased steadily as the fiber content increased due to poor bonding between the untreated fiber and polymer. Polyisocyanate was used as a coupling agent to improve tensile strength, but it had no effect on the modulus of the composites. Both untreated and isocyanate-treated composites had low impact strength values; further composite matrix modification would be necessary to maintain or improve impact strength.

Joseph et al (1993) investigated tensile properties of short sisal-reinforced polyethylene composites. Sisal is a ligonocellulosic like jute, coir, and pineapple, and it is of particular interest for use as a reinforcement in thermoset matrices, because it has high impact strength besides having moderate tensile and flexural properties compared to other lignocellulosic fibers. A preliminary investigation showed that sisal fiber could also be used as a reinforcement in a thermoplastic matrix and that it performed better than did wood fibers. The influence of the processing method and the effect of fiber content, fiber length, and orientation on tensile properties of the composite were evaluated. The tensile properties of the composites showed a gradual increase with fiber content. The properties also increased with fiber length, to a maximum at a fiber length of about 6 mm. Unidirectional alignment of the short fibers achieved by an extrusion process enhanced the tensile strength and modulus of the composites along the axis of fiber alignment by more than twofold, compared to randomly oriented fiber composites.

Karmaker et al (1994) studied the influence of water uptake on the mechanical properties of jute fiber-reinforced polypropylene. Being hydrophilic, jute fibers absorb a high amount of water, causing swelling of the fibers. As polypropylene left some gaps when it was shrunk by heat, the swelling of the jute fibers could fill these gaps. Then, it could give positive results on the mechanical properties of the composites and result in higher shear strength between fibers and matrix during fracture.

George et al (1995) studied composites of short pineapple leaf fiber reinforced low density polyethylene, which were prepared by two different methods, melt-mixing and solution-mixing methods. Tensile properties of melt-mixed and solution-mixed composites were compared. Solution-mixed composites showed better properties than melt-mixed composites. The influence of fiber length, fiber loading, and orientation on the mechanical properties was also evaluated. Considering the overall mechanical properties and processability characteristics, a fiber length of 6 mm was found to be the optimum length of pineapple leaf fiber for reinforcement in low density polyethylene (LDPE). The mechanical properties were enhanced and elongation at break reduced with increasing fiber loading. Longitudinally oriented composites showed better properties than randomly and transversely oriented composites. A comparison of the properties of the pineapple leaf fiber (PALF)-reinforced LDPE composites with those of other cellulose-fiber-reinforced LDPE system indicated superior performance of the PALF-LDPE composites.

Marcovich et al (1996) used sawdust, obtained from Eucaliptus saligna, or calcium carbonate as fillers, and combined them with an unsaturated polyester matrix. The ultimate strength, elongation, and water absorption decreased with the addition of fillers. On the other hand, Young's modulus was increased. The dynamic mechanical properties were used to determine the influence of the moisture content on the performance of the material.

Herrera-Franco and Aguilar-Vega (1997) studied the effect of cellulosic fiber, obtained from henequen (Agrave fourcroydes), combined with LDPE. The reinforcing fibers were used with three different types of surface treatment: without any treatment, preimpregnation with a solution containing 2% by weight of LDPE in xylene at 130 C under reflux for one hour conducted under a nitrogen atmosphere, and using a silane coupling agent applied at a concentration of 1% by weight in carbon tetrachloride at 70 C with dicumile peroxide as the catalyst. The concentration of randomly oriented fibers in the composite ranged between 0 and 30% by volume. The tensile strength and Young's modulus were increased with the addition of fibers. However, strain values were decreased, and the thermal behavior of the composite matrix was affected.

Preimpregnation of the cellulosics in a LDPE-xylene solution and the use of a silane coupling agent resulted in a small increment in the mechanical properties of the composites. The shear properties of the composites also increased with increasing of fiber content and fiber surface treatment. It was also found that the fiber surface treatment improves fiber dispersion in the matrix.

Devi et al (1997) used pineapple leaf fiber (PALF) as a polymer reinforcement because it is rich in cellulose, relatively inexpensive, and abundantly available. This study investigated the tensile, flexural, and impact behavior of PALF-reinforced polyester composites as a function of fiber loading, fiber length, and fiber surface modification. Increase in fiber content increased tensile strength and Young's modulus of the composites, as well as elongation at break. The mechanical properties were optimum at a fiber length of 30 mm. The specific flexural stiffness of the composite was about 2.3 times greater than that of neat polyester resin. The impact strength was increased with increase in fiber content, also. To improve the tensile strength, silane A172-treated fibers were combined into composites. The PALF polyester composite possessed superior mechanical properties compared to other cellulose-based natural fiber composites.

Oksman and Clemons (1998) studied the mechanical properties and morphology of impact modified polypropylene (PP)-wood fiber composites. PP has poor impact properties, especially at low temperatures. The impact properties were improved through the use of two different ethylene/propylene/diene terpolymers (EPPM) and one maleated styrene-ethylene/butylene-styrene triblock copolymer (SEBS-MA) as impact modifiers in the PP/wood fiber systems. All three elastomers increased the impact strength of the PP/wood fiber. Maleated polypropylene (MAPP) was used as a compatibilizer. Addition of MAPP alone did not affect the impact properties but it had a positive effect when used together with elastomers. In tensile tests, MAPP showed a negative effect on the elongation at break but a positive effect on tensile strength. MAPP further enhanced adhesion between wood flour and impact-modified PP systems. The impact modifiers were found to decrease the stiffness of the composites. Scanning electron microcopy showed that maleated EPDM and SEBS had a stronger affinity for wood surfaces than did the unmodified EPDM. The maleated elastomers were expected to form a flexible interface around the wood particles, giving the composites better impact strength.

CHAPTER 2

MATERIALS AND METHODS

Materials

The composite materials consisted of two parts, the polymer matrix and the reinforcement or filler.

1) Polymer matrix

High Density Polyethylene (HDPE) supplied by Paxon was chosen as the polymer matrix. HDPE resins were combined with paper fibers using a co-rotating twin screw extruder. The temperature of the extruder was set at 150°C in each zone in order to be higher than the HDPE melting point, which is between 130 and 135°C.

For other properties, HDPE has a glass transition temperature of -120°C, and the density is above 0.945 g/cc. Moreover, HDPE is highly crystalline, being between 65 and 90% crystalline.

As HDPE has a highly crystalline structure, it tends to have good stiffness, good moisture barrier properties, a high melting point and high tensile strength compared to some other kinds of plastics like low-density polyethylene (LDPE). HDPE is also a hydrophobic and non-polar thermoplastic.

2) Reinforcements or Fillers

There were two kinds of paper fibers supplied by Interfibe which were used as reinforcements in the composite materials.

a) Mixed paper fiber

b) High-grade deinked paper fiber

Mixed paper is paper of varied quality; included in this category are office waste, boxboard cuttings and mill wrappers. Deinked paper has been through a deinking process, resulting in paper fiber which is white and has more brightness than mixed paper fiber.

Normally, the structure of paper fiber is polar, hydrophilic fibers, while the structure of a polymer matrix like HDPE is hydrophobic. It is known that the hydrophilic cellulosic fibers have little adhesion to the hydrophobic thermoplastic matrix. It is also known that the high viscosity of the matrix during composite fabrication hinders the paper fiber impregnation and, therefore, results in poor fiber-matrix interaction (Herrera-Franco and Aguilar-Vega, 1997).

The fiber cannot get mixed very well without adding some kind of coupling agent, such as silane. The fiber has an influence on the properties of the composites. Especially, factors like fiber content, fiber length, fiber orientation and types of fibers influence the strength and properties of the composite, as will be detailed in the next chapter.

Methods

1) Processing of composites

The paper fibers and HDPE resins were combined by using a co-rotating twin screw extruder (Werner-Pfleiderer ZSK 30). The extruder was set as follows:

Compounding speed = 120 rpm

Compounding % load = 90 %

Discharge temperature = 150°C

The feed rate varied depending upon the ratio of paper fibers in the composite materials. The ratios of paper fibers used in the experiment were 0%, 10%, 20%, 30% and 40%. Loading greater than 40% could not be achieved because of the limited efficiency of the extruder.

The extruder has 6 processing zones. In each zone, the temperature was set at 150°C. The water as a coolant was run to maintain the temperature throughout the process. HDPE resins were fed into the extruder at zone 1. In zone 2, the resins were melted and passed to zone 3 for combining with the paper fibers. The reason why the fibers were fed in zone 3 is to reduce fiber damage and gain fiber distribution. In zone 4, 5, and 6, the fibers mixed with the HDPE completely. The extrudate emerged from zone 6, and was cut into approximately 6 inch sections.

The extruded materials were compressed into sheets by a Carver laboratory press compression molding machine, model M. The extrudates were assembled between two ch

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chrome plates and a frame. There were two sizes of frames, 15×15×0.25 cm and 12.7×12.7×0.3175 cm

. The first size of frame was used to prepare samples for the tensile property tests, whereas the second was used to prepare samples for the impact test and water absorption. Mylar sheets, made from polyethylene terephthalate (PET), were placed between the frame and chrome plates to minimize problems with sticking. This configuration was called a "sandwich".

The sandwich was placed between both platens where the temperature was set at 150°C, the recommended temperature for HDPE. Hydraulic pressure at 30,000 psi was used to press the sandwich for 5 minutes. Then, it was cooled to 50°C, using cooling water. After the composite sheets were set completely, they were cut into the desired shapes, such as a dumbbell shape for tensile testing and a rectangular shape (0.5×2.5×0.125 in) for Izod impact testing, and water absorption testing.

2) Tensile properties

Following ASTM D638-90, Tensile Standard Test Method for Tensile Properties of Plastics, the test specimens of reinforced composite were prepared in the dimensions of type I. The specimens were shaped by using Tensilkut equipment into the dumbbell shape.

An Instron testing machine, model no.4201, was used at the ambient conditions (23 °C, 50% RH), with the parameters as follows: load cell capacity of 1000 lbs, pre-load value of 5 lbs, and cross head speed of 0.5 in/min. The specimens were gripped on both ends by the cross heads. The results of tensile strength, % elongation, modulus of elasticity and yield strength were calculated as outlined in the standard.

3) Izod impact strength

Izod impact strength was determined following ASTM D256-90b, Standard Test Methods for Impact Resistance of Plastics and Electrical Insulating Materials. The specimens were cut in the dimension of $0.5 \times 2.5 \times 0.125$ in and notched by the TMI notching cutter. The angle of notch was 22 ½° ± ½° and the depth of notch was 0.1 inch, according to the standard. The specimens were tested by using a TMI 43-I IZOD impact tester with 5-lb pendulum. They were held as a vertical cantilever beam and were broken by a single swing of the pendulum. When the sample failed, it was necessary to consider its type of failue. The types of failures were classified following the ASTM standard, as type C (Complete break), H (Hinge break), PC (Partial break), or NB (Non-break). Finally, the machine reported the test data for all samples, including the mean and standard deviation.

4) Water absorption

To test water absorption, the method used was ASTM D570-81, Standard Test Method for Water Absorption of Plastics. The specimens were cut in the dimensions of 2.5×0.5 in, and sandpaper was used to smooth the edges of the specimens to prevent water absorption from the uneven surface.

The specimens were dried in an oven at 50±3°C for 24 hours, cooling them in the desicator, and then immediately weighing them to the nearest 0.001 g. Next, the conditioned specimens were placed in boiling distilled water for 2 hours, and they all were immersed completely under water. Finally, they were dried and weighed to the nearest

0.001 g immediately. The percentage of water absorption was calculated by the following equation.

$$\%\ \text{water absorption} = \frac{\text{gain in weight (g)} \times 100}{\text{conditioned wt. (g)}} \qquad (31)$$

5) Comparison of mechanical properties between paper fibers and wood fibers

Several former experiments concentrated on mechanical property testing of wood fiber reinforced thermoplastics. Paper fibers as the reinforcements were compared to wood fibers in composites when they were fed in the same ratio, 40%, and in the same kind of polymer matrix, HDPE.

As the results of the comparison were related to the structure of the fibers and composites, a scanning electron microscope (SEM) and other equipment were used to show the structure, dispersion, and adhesion of fibers and polymer matrix in the composite materials.

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CHAPTER 3

RESULTS AND DISCUSSION

Results - Tensile Strength

The results of tensile strength measurements are tabulated in Table 3, and graphed in Figure 1. Statistical analysis at 95% by the least significant difference method (LSD) showed that there was no significant difference between the unreinforced material and composites with 10% or 20% fiber content. However, the difference was significant at fiber contents of 30% and 40%, for both kinds of fibers.

In comparing the effects of the two different kinds of paper fibers in the composites, statistical analysis showed that there was no significant difference in the tensile strength.

As can be seen in Figure 1, tensile strength tended to decrease when the fibers were added to the composites. Thus, pure HDPE samples have the highest tensile strength.

(See Appendix A for data and Appendix B for statistical analysis)

Table 3. Results of Tensile Strength

Tensile strength		
Materials	Mean (PSI)	SD (PSI)
0%fiber	3882	576.8
10%fiber	3679	216.75
20%fiber	3553	107.76
30%fiber	3199	372.49
40%fiber	3059	230.81
10%fiber (deinked)	3710	187.24
20%fiber (deinked)	3532	110.81
30%fiber (deinked)	3251	166.51
40%fiber (deinked)	3262	213.8

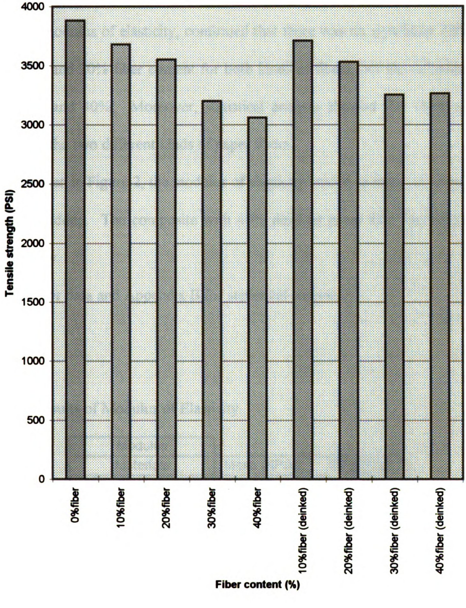

Figure 1. Tensile Strength

Results - Modulus of Elasticity

The results for modulus of elasticity are tabulated in Table 4 and presented graphically in Figure 2. Statistical analysis at 95% by LSD method, comparing the effect of paper fibers on modulus of elasticity, confirmed that there was no significant difference between 0%, 10%, and 20% fiber content for both kinds of fibers, but the difference was significant at 30% and 40%. Moreover, statistical analysis showed that there was no difference between the two different kinds of paper fibers.

As can be seen in Figure 2, the modulus of elasticity tended to increase when either kind of fibers was added. The composite with 40% deinked paper fiber had the highest modulus of elasticity.

(See Appendix A for data and Appendix B for statistical analysis)

Table 4. Results of Modulus of Elasticity

Modulus		
Materials	Mean (KPSI)	SD (KPSI)
0%fiber	110	47.89
10%fiber	127	37.76
20%fiber	192	74.42
30%fiber	223	72.48
40%fiber	347	74.68
10%fiber (deinked)	139	25.65
20%fiber (deinked)	177	16.23
30%fiber (deinked)	303	16.86
40%fiber (deinked)	402	81.13

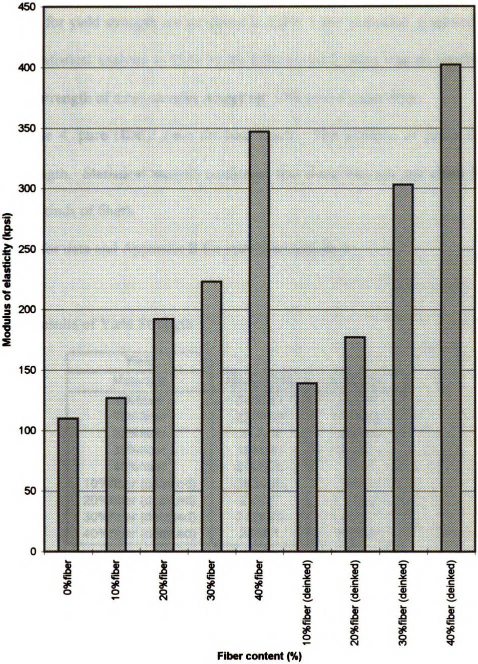

Figure 2. Modulus of Elasticity

Result - Yield Strength

The results for yield strength are tabulated in Table 5 and presented graphically in Figure 3. From statistical analysis at 95% by the LSD method, there was no significant difference in yield strength of most samples except for 30% mixed paper fiber.

From Figure 4, pure HDPE gave the best result. The addition of paper fibers decreased the strength. Statistical analysis confirmed that there was not any effect from using the different kinds of fibers.

(See Appendix A for data and Appendix B for statistical analysis)

Table 5. Results of Yield Strength

Yield		
Materials	Mean (PSI)	SD (PSI)
0%fiber	3850.11	571.56
10%fiber	2258.33	1240.33
20%fiber	2771.3	1082.59
30%fiber	1549.91	1084.6
40%fiber	2191.78	n/a
10%fiber (deinked)	3424.96	240.5
20%fiber (deinked)	2379.7	1279.56
30%fiber (deinked)	2409.75	1234.23
40%fiber (deinked)	2868.1	1014.38

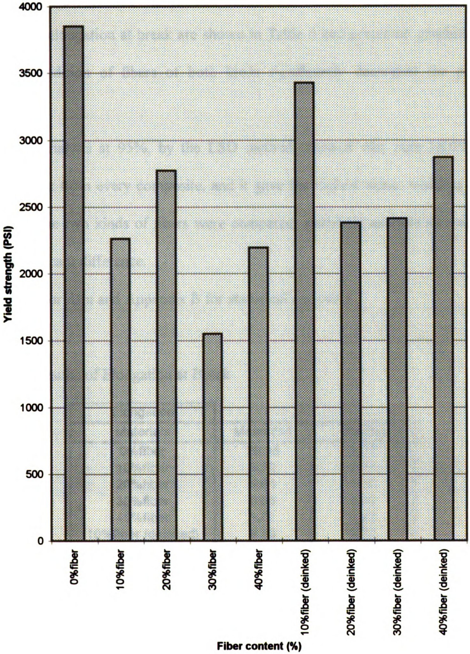

Figure 3. Yield Strength

Results - Elongation at Break

Results for elongation at break are shown in Table 6 and presented graphically in Figure 4. The addition of fibers of both kinds significantly decreased the percent elongation at break.

Statistical analysis at 95%, by the LSD method, showed that pure HDPE was statistically different from every composite, and it gave the highest value, which is about 26.96 %. When the two kinds of fibers were compared, statistical analysis showed that there was no significant difference.

(See Appendix A for data and Appendix B for statistical analysis)

Table 6. Results of Elongation at Break

Elongation		
Materials	Mean (%)	SD (%)
0%fiber	26.96	8.07
10%fiber	6.72	0.93
20%fiber	5.64	1.02
30%fiber	3.08	1.85
40%fiber	1.31	0.22
10%fiber (deinked)	8.55	0.31
20%fiber (deinked)	6.44	0.93
30%fiber (deinked)	2.44	0.66
40%fiber (deinked)	1.61	0.71

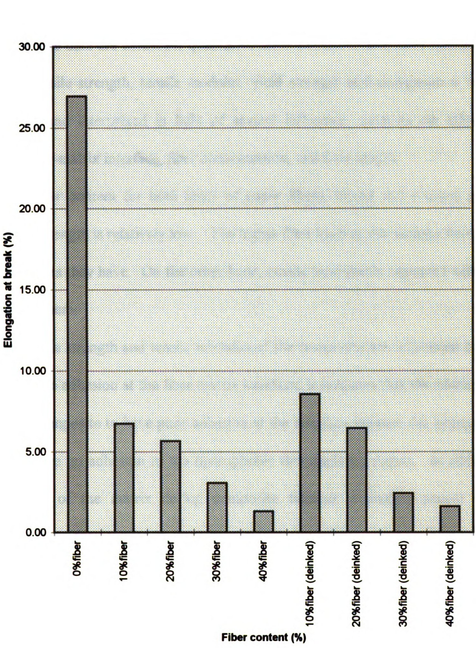

Figure 4. % Elongation at Break

Discussion - Tensile Properties

The results of tensile testing have been used to determine the mechanical strength of composites. These data are useful for qualitative characterization and development. In the experiment, tensile strength, tensile modulus, yield strength and elongation at break were determined, and interpreted in light of several influences, such as the effect of adhesion at the fiber-matrix interface, fiber concentration, and fiber length.

At 40% fiber content for both kinds of paper fibers, mixed and deinked paper fibers, the tensile strength is relatively low. The higher fiber loading the samples have, the lower strength values they have. On the other hand, tensile modulus is increased with the addition of paper fibers.

As the tensile strength and tensile modulus of the composite are influenced by the fiber loading and the adhesion at the fiber matrix interface, it indicates that the addition of fibers causes the composite to have poor adhesion at the interface because the hydrophilic cellulosic fibers have no adhesion to the hydrophobic thermoplastic matrix. In addition, the high viscosity of the matrix during composite fabrication hinders proper fiber impregation, and therefore results in poor-fiber matrix interaction. (Herrera-Franco and Aguilar-Vego,1997).

Yield strength and elongation have the same result as the tensile strength. The percentage of elongation of the paper fiber reinforced composites decreases with the increase in fiber loading. For example, at 40% fiber loading, the value of elongation at break was very low due to the poor interaction between fibers and the matrix, and it was statistically significantly different from the pure HDPE.

When the effects on tensile properties of the two kinds of paper fibers, at the same content in composites, were compared, statistical analysis showed no significant difference. However, the raw data for tensile strength suggested that the composite of deinked paper fibers had lower strength than that of mixed paper fibers. It is known that dieinking processes change the properties of paper fibers, besides removing ink. For example, the fiber length of deinked paper fiber is shorter than that of mixed paper fibers. As the length is shortened, the strength of the paper fibers is decreased.

As shown in Figures 5 to 8, the dispersion of the mixed and deinked paper fibers at 10% and 40% content in HDPE was investigated by SEM. The fibers were found to disperse randomly and not to be well mixed in the matrix. For instance, from Figure 6, at a magnification of 115 times, it can be seen that there is a bunch of dense fiber in one area, while few fibers are found in another area. This random dispersion contributes to the low strength of the composite.

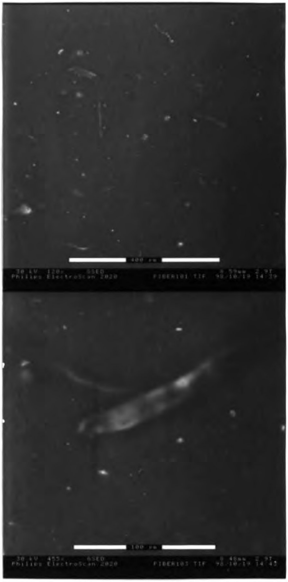

Figure 5. Dispersion of Mixed Paper Fiber Composites at 10%

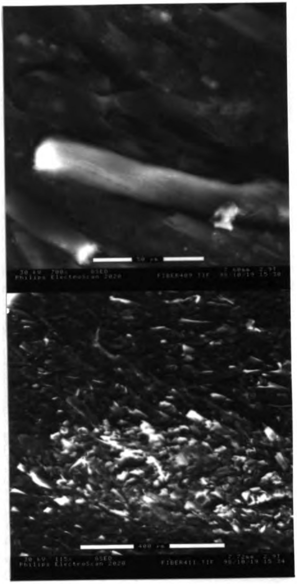

Figure 6. Dispersion of Mixed Paper Fiber Composites at 40%

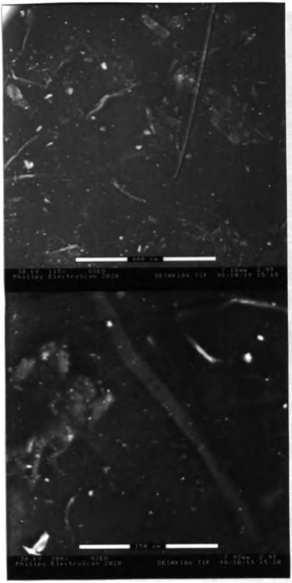

Figure 7. Dispersion of Deinked Paper Fiber Composites at 10%

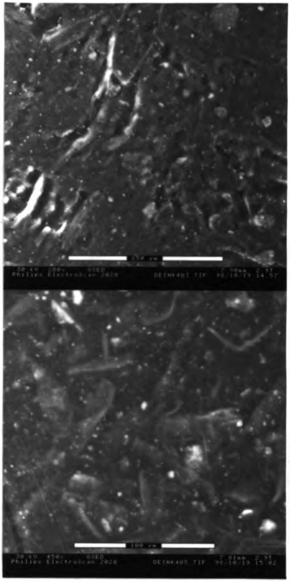

Figure 8. Dispersion of Deinked Paper Fiber Composites at 40%

Results - Izod Impact Strength

Results determined from Izod impact strength testing are summarized in Table 7 and presented graphically in Figure 9. They showed that the pure HDPE sample gave the best result in Izod impact strength, and it was statistically significantly different from the other samples. In addition, the characteristic type of break differed. Pure HDPE resulted in a partial break, whereas the other composites resulted in complete breaks.

From Figure 9, the addition of fibers of both kinds can be seen to have resulted in decreasing Izod impact strength. Statistical analysis at 95% by the LSD method confirmed that at 0% fiber, the result was statistically different from the samples with 10%, 20%, 30% and 40% fiber. However, there was no significant difference between the two kinds of fibers at the same content.

(See Appendix A for data and Appendix B for statistical analysis)

Table 7. Results of Izod Impact Strength

Materials	Mean (ftlb/in)	SD (ftlb/in)	Classify
0%fiber	1.730	0.108	Partial
10%fiber	0.653	0.023	Complete
20%fiber	0.604	0.051	Complete
30%fiber	0.488	0.048	Complete
40%fiber	0.407	0.065	Complete
10%fiber (deinked)	0.629	0.040	Complete
20%fiber (deinked)	0.556	0.022	Complete
30%fiber (deinked)	0.468	0.025	Complete
40%fiber (deinked)	0.368	0.062	Complete

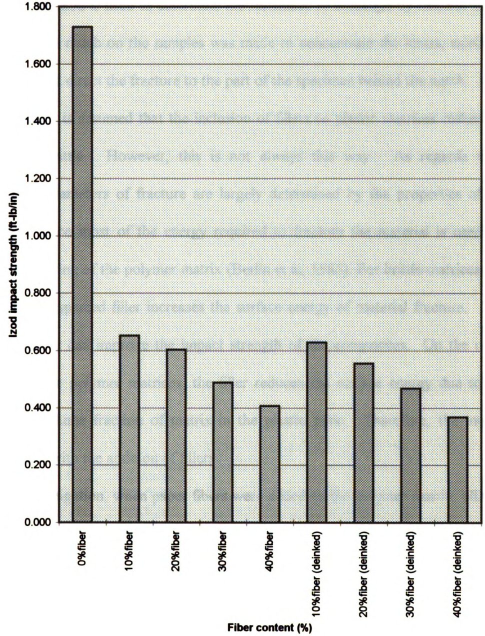

Figure 9. Izod Impact Strength

Discussion - Izod Impact Strength

This test method is used to determine the resistance to breakage by flexural shock of composites. The notch on the samples was made to concentrate the stress, minimize the deformation, and direct the fracture to the part of the specimen behind the notch.

Generally, it is assumed that the inclusion of fillers to plastic matrices makes the composite more brittle. However, this is not always this way. As regards filled composites, the parameters of fracture are largely determined by the properties of the polymer matrix, since most of the energy required to fracture the material is used for straining and fracturing of the polymer matrix (Berlin et al, 1986). For brittle matrices, the introduction of a dispersed filler increases the surface energy of material fracture. This means that the filler can improve the impact strength of the composites. On the other hand, for non-brittle polymer matrices, the filler reduces the surface energy due to the decrease of the volume fraction of matrix in the plastic zone. Therefore, the impact strength decreases with the addition of fillers.

In this investigation, when paper fibers were added to the polymer matrix, HDPE, the impact strength decreased due to the ductile nature of HDPE. This means that the paper fibers make the composites more brittle. Statistical analysis showed a significant difference in the results when fibers were added.

Results - Water Absorption

The results of water absorption are tabulated in Table 8 and presented graphically in Figure 12. Statistical analysis at 95% by LSD method confirmed that there was a significant difference among the samples, which had different content of paper fibers. From Figure 12, it can be seen that the addition of the fibers increased the percentage of water absorption for both kinds of paper fibers. At 40% mixed paper fiber, the water absorbed, was highest.

When comparing the effect of using different kinds of paper fibers, mixed and deinked paper fibers, statistical analysis showed a significant difference between pairs of samples at the same percentage of paper fibers, except at 10% fiber.

(See Appendix A for data and Appendix B for statistical analysis)

Table 8. Results of Water Absorption

Materials	mean (%)	SD (%)
0%fiber	0	0
10%fiber	0.090746	0.015206
20%fiber	0.214712	0.020299
30%fiber	0.229	0.010234
40%fiber	0.717783	0.053387
10%fiber (deinked)	0.066394	0.003862
20% fiber (deinked)	0.15311	0.005326
30% fiber (deinked)	0.51812	0.028953
40% fiber (deinked)	0.621157	0.031183

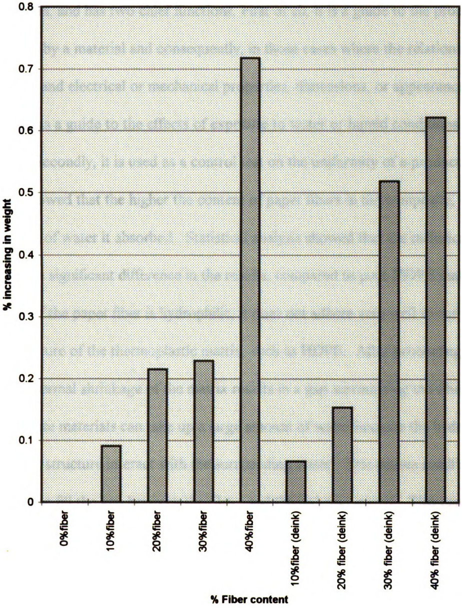

Figure 10. Water Absorption

Discussion - Water absorption

This test method, following ASTM D 570-81, is used to determine the relative rate of water absorption, and has two chief functions. First of all, it is a guide to the proportion of water absorbed by a material and consequently, in those cases where the relationships between moisture and electrical or mechanical properties, dimensions, or appearance have been determined, as a guide to the effects of exposure to water or humid conditions on such properties. Secondly, it is used as a control test on the uniformity of a product.

Results showed that the higher the content of paper fibers in the composite, the higher the amount of water it absorbed. Statistical analysis showed that the inclusion of paper fibers gave a significant difference in the results, compared to pure HDPE materials. As the structure of the paper fiber is hydrophilic, it does not adhere very well to the hydrophobic structure of the thermoplastic matrix, such as HDPE. After processing of the composites, the thermal shrinkage of the matrix results in a gap surrounding the fiber. Then, the composite materials can take up a large amount of water because the hydroxyl groups in the fiber structure interact with the surrounding water. This causes swelling of the fibers which can fill the gap between the fibers and the polymer matrix. This can result in a decrease in mechanical properties.

The higher the amount of fibers the composite has, the more gaps can occur. Thus, at 40% fiber, the water is absorbed easily compared to pure HDPE, and to composites, with lower contents of fibers.

When comparing how well the different kinds of fibers can absorb water, it seems that mixed paper fibers can absorb a greater amount of water than the deinked paper fibers (except at 30% fiber). Biermann (1993) explained that to disperse ink during the deinking process, it requires using a vehicle, which is commonly vegetable oil. The effect of the oil, which is a hydrophobic structure, would cause a partial reduction in the water absorption efficiency of the fibers.

Results - Comparison of wood fiber (Aspen fibers) and paper fiber (Mixed and deinked paper fibers) reinforced HDPE composites (at 40% fiber and 60% HDPE)

Paper fiber-reinforced HDPE composites were compared to wood fiber-reinforced HDPE composites from prior experiments by Childress (1991). To investigate the properties of composites, she used aspen hardwood fiber combined with HDPE. Several kinds of additives were used to improve the properties. In this case, the composite of 40% aspen without using any additives was selected to compare to the composites with 40% of both paper fibers. Moreover, they all were compared to the properties of pure HDPE. The tensile modulus of the wood fiber composites is not so much different from that of the HDPE materials, but lower than that of the paper fiber composites. Decreases in the tensile strength, % elongation at break, and Izod impact strength resulted with the addition of the fibers, so the HDPE samples were expected to have the highest values of all properties, whereas the paper fiber composites gave the lowest values. This means that the composites of the paper fibers could be the most brittle materials, and the HDPE could be the toughest materials. For water absorption, the wood fiber composites absorbed more water than the paper fiber composites, whereas the pure HDPE did not absorb water at all.

From Figures 11, 12, and 13, a scanning electron microscope (SEM) was used to show the structures of mixed paper fibers, deinked paper fibers, and aspen wood fibers. As can be seen, both kinds of paper fiber structures are similar. They are flat and entangled, whereas the aspen fibers quite look quite straight. The fiber length was compared among the three kinds of fibers, as follows:

Mixed paper fibers	:	450 μm < Length < 1mm
Deinked paper fibers	:	Length < 450 μm
Aspen wood fibers	:	Length > 1 mm

Table 9 Comparison of Properties of Wood and Paper Fiber Composites

	HDPE	Aspen fiber	Mixed paper fiber	Deinked paper fiber
Properties		at 40% with 60% HDPE in composites		
Tensile strength (PSI)	3882	2929.9	3059	3262
Tensile modulus (KPSI)	110	114.73	347	241
% Elongation at break	26.96	3.81	1.31	1.61
Izod impact strength (ftlb/in)	1.730	0.98	0.407	0.368
% Water absorption	0	2.11	0.71	0.62

Discussion - Comparison of Properties of Wood and Paper Fiber Composites

Childress (1991) described the natural characteristics of aspen fibers as polar in nature and hydrophilic. In the processing, the fibers were in the form of thermomechanical pulp (TMP). This mechanical pulping process is one in which the fibers retain most of their lignin and natural waxes. Aspen wood fiber length is typically 0.7 - 3 mm.

For paper fibers, they are also polar and hydrophilic. In the processing, both kinds of papers, mixed and deinked paper fibers, pass through a pulping system to remove contaminants, such as plastic laminates, adhesives, waxes, etc. However, there is a difference in processing between the paper fibers: deinked paper fibers also have to pass through the deinking process to remove ink in order to get good printing quality.

The effects of both paper fibers on physical properties were already discussed. Therefore, at this point, only the effects of aspen wood fibers compared to paper fibers on physical properties will be explained.

The significant difference between wood and paper fibers is the fiber length. George et al (1995) explained that the strength of fiber-reinforced composites depends on the degree to which an applied load is transmitted, and the extent of load transmittance is a function of fiber length and magnitude of the fiber-matrix interfacial bond. The short fibers will debond from the matrix easily and the composite will fail at a low load. When the fiber length is long, the stressed composites will lead to breaking of fibers and a high composite strength. Figures 11, 12, and 13 show SEM photographs of the wood and paper fibers. As can be seen, aspen wood fibers are much longer in length than paper fibers, so the strength of the wood fiber composites is expected to be higher.

As can be seen in Table 9, the paper fiber composites have high tensile strength, and their stiffness is improved as compared to the aspen wood fiber composite and HDPE materials. Their water absorption decreases. The only loss is the impact strength, which they are expected to reduce. For the wood fiber composites, the tensile strength is as least as good, and they are expected to be more brittle and stiffer in comparison to the HDPE materials. The wood fiber composites are the best for absorbing the water.

Smook (1992) explained the effect of recycling of the wastepaper on the fiber strength and on the bonding strength between fibers. The strength decreases and the bonding between fibers shows a more dramatic loss. These factors can support why paper fiber composites have lower strength than wood fiber composites.

For % water absorption, Smook (1992) said that, in recycling processes, with each drying and slushing cycle, the fibers become less flexible and less permeable to water, and therefore do not conform as well as virgin fibers. Thus, aspen wood fibers can absorb water better than paper fibers.

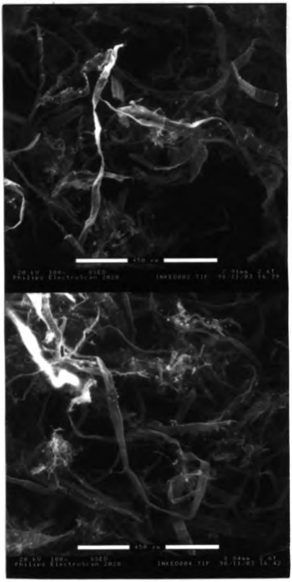

Figure 13. Mixed Paper Fibers

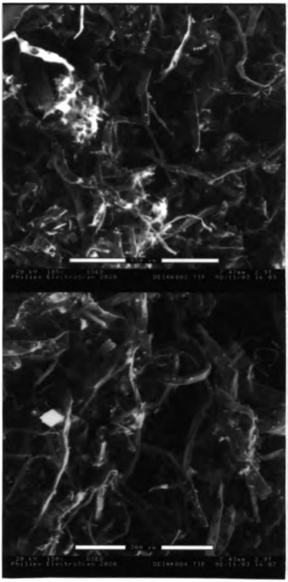

Figure 14. Deinked Paper Fibers

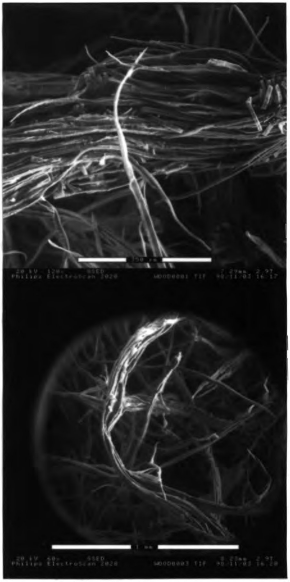

Figure 15. Aspen Wood Fibers

Summary and Conclusions

Paper fiber reinforced HDPE composites were prepared by the twin-screw extruder and compression molding at ratios of 0%, 10%, 20%, 30% , and 40% fiber. Mixed and deinked paper fibers were used as fillers to determine whether the deinking process had an effect on the mechanical properties of the composite materials.

Properties of fiber-reinforced composites depend on many factors like fiber-matrix adhesion, volume fraction of fiber, fiber aspect ratio, and fiber orientation as well as the stress transfer efficiency of the interface.

Mechanical properties of composites were studied which included tensile stress, tensile modulus, yield strength, and % elongation. Tensile modulus increased when either mixed or deinked paper fibers were added. The worst result for composite strength was at 40% fiber which was due to the low values for tensile strength and % elongation at break.

Izod impact strength of the composites decreased greatly, as is expected for tough materials such as HDPE. Water absorption increased with increasing fiber content.

Statistical analysis of the properties showed little effect of the kind of the fiber on the properties of the composites. However, the deinking process can reduce the fiber length of deinked paper fibers and deinked paper fibers will be shorter than the length of mixed paper fibers, as can be seen by the SEM photographs shown in Figures 11 and 12. Because the length of deinked paper fiber is just slightly shorter, so its strength is a little bit weaker than the strength of mixed paper fibers.

When the properties were compared to composites of aspen wood fibers, the paper fiber composites gave poorer results in terms of the tensile modulus, % elongation at break, and Izod impact strength because they were more brittle and easier to break.

This method is a potential way to reduce the amount of waste paper, which can cause a lot of problems in the environment. Even though a high amount of paper fibers can be used to make composite materials, which will increase the utilization rate for recycled papers, composite properties are reduced because of the lack of interfacial bonding between the HDPE matrices and the paper fibers, due to the difference of their potential bonding properties, hydrophobic and hydrophilic. If the bonding was improved, the properties of paper fiber-reinforced HDPE composites would be better.

Recommendation for Future Research

As the incompatibility of the fibers and the matrix, including poor dispersion of the fibers in the matrix, causes adhesion between them in the composites to be relatively poor, so it causes the mechanical and physical properties to have poor results.

To improve the bonding at the interface, coupling agents should be investigated to improve dispersion, adhesion, and compatibility for a system containing hydrophilic cellulose and a hydrophobic matrix.

Moreover, there are several other kinds of paper fibers to be investigated for use in the composite as a filler. If this can be successful, it will be a great opportunity to increase paper recycling in the future.

APPENDIX A

Table 10. Tensile Strength Data

Tensile strength (PSI)							
Materials	1	2	3	4	5	Mean	SD
0%fiber	2867	4177	4002	4281	4083	3882	576.8
10%fiber	3719	3969	3753	3564	3390	3679	216.75
20%fiber	3539	3655	3673	3459	3441	3553	107.76
30%fiber	2882	2990	3287	3810	3026	3199	372.49
40%fiber	2798	2986	3343	3254	2915	3059	230.81
10%fiber deink	3976	3776	3734	3558	3507	3710	187.24
20%fiber deink	3594	3527	3383	3674	3480	3532	110.81
30%fiber deink	3149	3366	3008	3338	3395	3251	166.51
40%fiber deink	3063	3006	3467	3432	3344	3262	213.8

Table 11. Modulus of Elasticity Data

Modulus of Elasticity (KPSI)							
Materials	1	2	3	4	5	Mean	SD
0%fiber	166	157	62	82	83	110	47.89
10%fiber	176	89	155	120	94	127	37.76
20%fiber	112	229	281	117	219	192	74.42
30%fiber	318	285	171	165	176	223	72.48
40%fiber	317	379	422	384	232	347	74.68
10%fiber (deinked)	166	142	107	119	161	139	25.65
20%fiber (deinked)	185	150	172	184	192	177	16.23
30%fiber (deinked)	298	304	330	296	285	303	16.86
40%fiber (deinked)	n/a	388	461	292	465	402	81.13

Table 12. Yield Strength Data

Yield Strength (PSI)							
Materials	1	2	3	4	5	Mean	SD
0%fiber	2852.76	4173.9	3993.71	4267	3963.17	3850.11	571.56
10%fiber	2982.83	828.36	988.79	3350.51	3141.15	2258.33	1240.33
20%fiber	3347.86	3488.48	875.5	2890.77	3253.88	2771.3	1082.59
30%fiber	858.39	n/a	3168.76	1097.01	1075.49	1549.91	1084.6
40%fiber	1077.44	n/a	3306.12	n/a	n/a	2191.78	n/a
10%fiber (deinked)	3830.3	3452.95	3233.05	3316.69	3291.83	3424.96	240.5
20%fiber (deinked)	3388.39	3254.28	3297.43	1008.95	949.48	2379.7	1279.56
30%fiber (deinked)	1063.78	3340.85	1054.35	3221.53	3368.21	2409.75	1234.23
40%fiber (deinked)	n/a	1351.28	3463.69	3388.26	3269.19	2868.1	1014.38

Table 13. Elongation at Break Data

Elongation at break (%)							
Materials	1	2	3	4	5	Mean	SD
0%fiber	19.76	20.76	30.47	24.52	39.29	26.96	8.07
10%fiber	5.65	7.7	5.95	6.72	7.6	6.72	0.93
20%fiber	6.89	5.38	4.14	6.14	5.66	5.64	1.02
30%fiber	1.44	2.09	4.79	5.35	1.71	3.08	1.85
40%fiber	1.13	1.27	1.53	1.08	1.54	1.31	0.22
10%fiber (deinked)	8.13	8.43	8.85	8.85	8.5	8.55	0.31
20%fiber (deinked)	7.05	7.67	6	6.2	5.3	6.44	0.93
30%fiber (deinked)	2.36	3.12	1.4	2.86	2.45	2.44	0.66
40%fiber (deinked)	1.17	0.83	1.38	2.52	2.17	1.61	0.71

Table 14. Izod Impact Strength Data (ft-lb/in)

Materials	1	2	3	4	5	Mean	SD
0%fiber	1.609	1.86	1.635	1.808	1.737	1.730	0.108
10%fiber	0.677	0.635	0.657	0.625	0.672	0.653	0.023
20%fiber	0.683	0.583	0.613	0.597	0.544	0.604	0.051
30%fiber	0.538	0.477	0.534	0.469	0.423	0.488	0.048
40%fiber	0.394	0.522	0.372	0.366	0.382	0.407	0.065
10%fiber (deinked)	0.662	0.597	0.678	0.622	0.588	0.629	0.040
20%fiber (deinked)	0.555	0.542	0.588	0.564	0.531	0.556	0.022
30%fiber (deinked)	0.433	0.488	0.458	0.494	0.469	0.468	0.025
40%fiber (deinked)	0.403	0.413	0.423	0.3	0.3	0.368	0.062

Table 15. Water Absorption

	weight before absorbing (lb)			weight after absorbing (lb)			different in weight				
Materials	1	2	3	1	2	3	1 (lb)	2 (lb)	3 (lb)	mean(%)	SD(%)
0%fiber	0	0	0	0	0	0	0	0	0	0	0
10%fiber	2.5404	2.4428	2.5035	2.5431	2.445	2.5054	0.106282	0.090061	0.075894	0.090746	0.015206
20%fiber	2.7	2.8604	2.6485	2.7054	2.8663	2.6548	0.2	0.206265	0.23787	0.214712	0.020299
30%fiber	2.8482	2.9888	2.9759	2.8544	2.9959	2.9828	0.217681	0.237554	0.231863	0.229033	0.010234
40%fiber	3.121	3.0112	2.8927	3.1449	3.0331	2.9118	0.76578	0.727285	0.660283	0.717783	0.053387
10%fiber (deinked)	2.5358	2.5703	2.5754	2.5375	2.5719	2.5772	0.06704	0.06225	0.069892	0.066394	0.003862
20% fiber (deinked)	2.6365	2.789	2.7421	2.6406	2.7931	2.7464	0.155509	0.147006	0.156814	0.15311	0.005326
30% fiber (deinked)	2.9622	2.9103	2.8755	2.9766	2.9256	2.8911	0.486125	0.525719	0.542514	0.51812	0.028953
40% fiber (deinked)	1.879	1.802	1.9228	1.8913	1.8131	1.9342	0.654604	0.615982	0.592885	0.621157	0.031183

APPENDIX B

One-Way Analysis of Variance of Tensile Strength

Data: Tensile Strength

Level codes: Treatment
Means plot: LSD *Confidence level: 95* *Range test: LSD*

Analysis of variance

Source of variation	Sum of Squares	d.f.	Mean square	F-ratio	Sig. level
Between groups	3069420.0	8	383677.50	4.904	.0004
Within groups	2816290.0	36	78230.28		
Total (corrected)	5885710.0	44			

0 missing value(s) have been excluded.

Multiple range analysis for Tensile Strength

Method: 95 Percent LSD

Level	Count	Average	Homogeneous Groups
40% fiber	5	3059.2000	X
30% fiber	5	3199.0000	XX
30% deink fiber	5	3251.2000	XX
40% deink fiber	5	3262.4000	XX
20% deink fiber	5	3531.6000	XX
20% fiber	5	3553.4000	XX
10% fiber	5	3679.0000	X
10% deink fiber	5	3710.2000	X
0% fiber	5	3882.0000	X

One-Way Analysis of Variance of Elongation

Data: Elongation
Level codes: Treatment
Means plot: LSD   *Confidence level: 95*   *Range test: LSD*

Analysis of variance

Source of variation	Sum of Squares	d.f.	Mean square	F-ratio	Sig. level
Between groups	2503.1938	8	312.89923	38.898	.0000
Within groups	289.5901	36	8.04417		
Total (corrected)	2792.7839	44			

0 missing value(s) have been excluded.

Multiple range analysis for Elongation

Method: 95 Percent LSD

Level	Count	Average	Homogeneous Groups
40% fiber	5	1.3100000	X
40% deink fiber	5	1.6140000	X
30% deink fiber	5	2.4380000	XX
30% fiber	5	3.0760000	XXX
20% fiber	5	5.6420000	XXX
20% deink fiber	5	6.4440000	XX
10% fiber	5	6.7240000	X
10% deink fiber	5	8.5520000	X
0% fiber	5	26.960000	X

One-Way Analysis of Variance of Yield

Data: Yield
Level codes: Treatment
Means plot: LSD *Confidence level: 95* *Range test: LSD*

Analysis of variance

Source of variation	Sum of Squares	d.f.	Mean square	F-ratio	Sig. level
Between groups	17091387	8	2136423.4	1.941	.0889
Within groups	34121633	31	1100697.8		
Total (corrected)	51213020	39			

3 missing value(s) have been excluded.

Multiple range analysis for Yield

Method: 95 Percent LSD

Level	Count	Average	Homogeneous Groups
30% fiber	4	1549.9125	X
40% fiber	2	2191.7800	XX
10% fiber	5	2258.3280	XXX
20%deink fiber	5	2379.7060	XXX
30%deink fiber	5	2409.7440	XXX
20% fiber	5	2771.2980	XXXX
40% deink fiber	4	2868.1050	XXXX
10% deink fiber	5	3424.9640	XXX
0% fiber	5	3850.1080	X X

One-Way Analysis of Variance of Modulus of Elasticity

Data: Modulus of Elasticity
Level codes: Treatment
Means plot: LSD *Confidence level: 95* *Range test: LSD*

Analysis of variance

Source of variation	Sum of Squares	d.f.	Mean square	F-ratio	Sig. level
Between groups	396504.35	8	49563.043	16.541	.0000
Within groups	104870.20	35	2996.291		
Total (corrected)	501374.55	43			

1 missing value(s) have been excluded.

Multiple range analysis for Modulus of Elasticity

Method: 95 Percent LSD

Level	Count	Average	Homogeneous Groups
0% fiber	5	110.00000	X
10%fiber	5	126.80000	XX
10%deink fiber	5	139.00000	XX
20%deink fiber	5	176.60000	XXX
20% fiber	5	191.60000	XX
30% fiber	5	223.00000	X
30%deink fiber	5	302.60000	X
40% fiber	5	346.80000	XX
40%deink fiber	4	401.50000	X

One-Way Analysis of Variance of Impact Strength

Data: Impact Strength
Level codes: Treatment
Means plot: LSD *Confidence level: 95* *Range test: LSD*

Source of variation	Sum of Squares	d.f.	Mean square	F-ratio	Sig. level
Between groups	6.8738856	8	.8592357	277.430	.0000
Within groups	.1114964	36	.0030971		
Total (corrected)	6.9853820	44			

0 missing value(s) have been excluded.

Multiple range analysis for Impact Strength
Method: 95 Percent LSD

Level	Count	Average	Homogeneous Groups
40% deink fiber	5	.3678000	X
40% fiber	5	.4072000	XX
30% deink fiber	5	.4684000	XX
30% fiber	5	.4882000	XX
20% deink fiber	5	.5560000	XX
20% fiber	5	.6040000	XX
10%deink fiber	5	.6294000	X
10% fiber	5	.6532000	X
0% fiber	5	1.7298000	X

One-Way Analysis of Water Absorption (%)

Data: Water Absorption (%)
Level codes: Treatment
Means plot: LSD *Confidence level: 95* *Range test: LSD*

Analysis of variance

Source of variation	Sum of Squares	d.f.	Mean square	F-ratio	Sig. level
Between groups	1.6398826	8	.2049853	338.381	.0000
Within groups	.0109041	18	.0006058		
Total (corrected)	1.6507866	26			

18 missing value(s) have been excluded.

Multiple range analysis for Water absorption

Method: 95 Percent LSD

Level	Count	Average	Homogeneous Groups
0% fiber	3	.0000000	X
10%deink fiber	3	.0663940	X
10% fiber	3	.0907457	X
20%deink fiber	3	.1531097	X
20% fiber	3	.2147117	X
30% fiber	3	.2290327	X
30%deink fiber	3	.5181193	X
40%deink fiber	3	.6211570	X
40% fiber	3	.7177827	X

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