CHAPTER 3

DEVELOPMENT OF NATURAL FIBER REINFORCED THERMOPLASTIC

The development of the natural fiber reinforced thermoplastic composite involves four stages of processes. The first process is the natural fiber processing and made into roving. The natural fibers like kenaf, sisal and jute undergoes several processes before they are ready to be used in the fiber reinforced composites. The second stage of this study is the long fiber Pellet fabrication. The hot impregnation process is used to prepare the long fiber pellets with pp matrix. The third stage is the injection moulded specimens fabricated according to standards. The last stage is the testing of the specimens for mechanical, thermal, and recycling properties.

3.1 PROCESSING OF NATURAL FIBERS

3.1.1 Jute Fiber Processing and Roving

- The extraction of the natural fiber from the plant required certain care to avoid damage.

- In the present experiments, initially the jute sections were cut from the main stem of the plant and then rolled lightly to remove the excess moisture.
Impurities in the rolled fibers such as pigments, broken fibers, coating of cellulose etc., were removed manually by means of comb, and then the fibers were cleaned and dried.

This was followed by cleaning and drying of the fibers in a chamber at 200° C for three hours. Figure 3.1 shows the processing of the fibers in the oven.

### 3.1.2 Sisal Fiber Extraction

- Sisal Fiber is extracted from the leaves of Sisal Plant. The fibers are extracted through hand extraction machine composed of either serrated or non serrated knives.
- The peel is clamped between the wood plank and knife and hand-pulled through, removing the resinous material.
- The extracted fibers are sun-dried for whitening them. Once dried, the fibers are ready for knotting. A bunch of fibers is mounted or clamped on a stick to facilitate segregation.
- Each fiber is separated according to fiber sizes and grouped accordingly.
- To knot the fiber, each fiber is separated and knotted to the end of another fiber manually.

### 3.1.3 Kenaf Fiber Extraction

- Kenaf Fiber is extracted from the bast of kenaf Plant. The fibers are extracted through hand extraction machine composed of either serrated or non serrated knives.
The average length of the core fibers range is from 0.49 to 0.78 mm long with a mean length of 0.6 mm and average diameter of 37.4 mm. The core pulp, compared to hardwood pulps, has lower tear strength, but greater tensile and burst strength.

A bunch of fibers are mounted or clamped on a stick to facilitate segregation. Each fiber is separated according to fiber sizes and grouped accordingly.

To make the rope, the lengthy fibers are towed into rovings.

Figure 3.1 Processing of fibers in oven for removing moisture

3.2 THE EFFECT OF ALKALI CHEMICAL TREATMENT IN THE FIBERS

Chemical treatment was applied to kenaf, sisal and jute natural fibers to create better fiber resin adhesion in natural fibers. The natural fibers
have been treated with varying concentrations of caustic soda with the objective of removing surface impurities and developing fine structure modifications in the process of mercerisation. Kenaf, sisal and jute fibers were taken in separate trays; to these trays 10% NaOH solution was added, and the fibers were soaked in the solution for 10 hours (figure 3.2). The fibers were then washed thoroughly with water to remove the excess of NaOH sticking to the fibers.

![Figure 3.2 Soaking of fibers in NaOH solution](image)

3.3 HOT IMPREGNATION PROCESS

The raw materials used in this research were

**Polymer:** Commercial grade PP

**Reinforcement Fibers:** kenaf, jute, sisal fiber

**Compatibilizer:** Maleated Polypropylene (MAPP)
The ratio of the matrix and the fiber loading is important and the optimized percentage were referred in the literature review (Suhara et al, 2006). In that experiment the 40% of the fiber to the PP matrix contributed the better results compared to 30% and 50% fiber loading.

Formulation of composites used in this research has abbreviations as follows:-

1. 40% wt of twisted kenaf fiber + polypropylene + compatibilizer; KLFRT.
2. 40% wt of twisted jute fiber + polypropylene + compatibilizer; JLFRT.
3. 40% wt of twisted sisal fiber + polypropylene + compatibilizer; SLFRT.
4. 40% long glass fiber filled, (GF-PP), which is available commercially; LFRT.

Figure 3.3  Schematic diagram of manufacturing process of the natural fiber reinforced composites
Alkali Chemical treatment with NaOH removes the moisture content from the fibers, thereby increasing its strength. Chemical treatment also enhances the flexural rigidity of the fibers. This treatment clears all the impurities in the fiber material and also stabilizes the molecular orientation; a hot melt impregnation process was used to fabricate impregnated natural fiber pellets. The entire manufacturing process of natural long fiber thermoplastic composite is detailed in Figures 3.3, 3.4 and the experimental setup is shown in Figure 3.5(a-d).

**Figure 3.4 Block diagram of hot impregnation process**

In this impregnation process kenaf, jute and sisal fiber rovings were used instead of synthetic glass roving. The natural kenaf fiber tows were sent along through the die to make the pp impregnated matrix over the natural fiber. Tows were protruded with natural fiber through a heated die during which the individual filaments were coated with matrix pp. This particular die unit was fitted as an accessory unit in the normal screw extruder, instead of pouring the fibers in the hoppers the fibers were towed and made as a roving and passed through the die. In the die the hot melt pp matrix formed a coating over the fiber which was then pelletized. The pultruded tow impregnated with the pp matrix was cooled and then chopped into pellets approximately 10 to 11 mm in length and 3 mm diameter. The molding conditions were listed in the Table 3.1. This molding conditions are very essential for the perfect mixing and matrix of the fiber and pp.
For this research four different composites (KLFRT, JLFRT, SLFRT, LFRT) were cut as pellets and used as a starting material for injection moulding process. The specimens were moulded according to the ASTM standards using injection molding process. The orientation of fibers in the course of action is anistropic and the flow axis is longitudinal. The length of the pellet is equal to length of the fiber in the pallets.

Figure 3.5 Manufacturing process of natural fiber composites
Table 3.1 Moulding conditions for natural long fiber composites during impregnation process

<table>
<thead>
<tr>
<th>Processing parameters</th>
<th>Set up value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mould temperature[^°C]</td>
<td>120</td>
</tr>
<tr>
<td>Melt temperature[^°C]</td>
<td>300</td>
</tr>
<tr>
<td>Volume flow rate [cm3/s]</td>
<td>80</td>
</tr>
<tr>
<td>Holding pressure [bar]</td>
<td>440</td>
</tr>
<tr>
<td>Back pressure [bar]</td>
<td>8</td>
</tr>
<tr>
<td>Screw rotation speed [rpm]</td>
<td>60</td>
</tr>
</tbody>
</table>

Figure 3.6 Injection molding process

Injection moulding is a process of forming an article by forcing molten plastic material under pressure into a mould where it is cooled, solidified and subsequently released by opening the two halves of the mould. Injection moulding is used for the formation of intricate plastic parts with excellent dimensional accuracy. Granules of natural fiber thermoplastic composite pallets were poured or fed into a hopper which stores it until it is needed. It is shown in Figure 3.6 (a-b). Due to heater action in the moulding
unit, the material is converted into liquid. Here the heater contains many layers where each layer is to be maintained at a particular temperature. Because of higher pressure the molten material is forced into the mould through the nozzle. Then this moulded material is converted into favourable ASTM (American society for testing and materials) sample.

3.4 TESTING OF SPECIMENS FOR MECHANICAL PROPERTIES

Density and mechanical properties such as tensile strength, tensile modulus, flexural strength, flexural modulus, impact strength were tested according to the ASTM standards. For Density – ASTM D 792, tensile properties – ASTM D3039, Flexural properties – ASTM D790, Impact strength – ASTM D 256,Dynamic mechanical analysis(DMA), Water absorption test, thickness swelling test were carried out with five set of specimens of KLFRT, JLFRT,SLFRT and LFRT. The values were compared with commercial LFRT composites.

3.4.1 Tensile Test

The ability to resist breaking under tensile stress is one of the most important and widely measured properties of materials used in structural applications. The force per unit area (MPa or psi) required to break a material in such a manner is the ultimate tensile strength or tensile strength at break.

Tensile properties indicate how the material will react to forces being applied in tension. A tensile test is a fundamental mechanical test where a carefully prepared specimen is loaded in a very controlled manner while measuring the applied load and the elongation of the specimen over some distance. Tensile tests are used to determine the modulus of elasticity, elastic limit, elongation, proportional limit, and reduction in area, tensile strength,
yield point, yield strength and other tensile properties. Figure 3.7(a) shows the specimen placed in the tensile testing machine to calculate the strength and modulus. Figure 3.7(b) and (c) shows the tensile test specimens before and after testing. The stress-strain curve relates the applied stress to the resulting strain and each material has its own unique stress-strain curve.

(a) Tensile specimen fixed in apparatus

(b) Specimen before testing (c) Specimen after testing

Figure 3.7 Tensile testing
Calculation for tensile strength and modulus

Tensile strength = $W / bd$

where,

$W$ is the ultimate failure load (N),

$b$ is mean width of sample (mm),

$d$ is mean thickness of sample (mm),

and $m$ is slope of stress-strain curve

For Polypropylene

Tensile strength = $W/bd$

$$= 290/(12.30 \times 3.5)$$

$$= 6.736 \text{MPa}$$

Tensile modulus = $m / bd$

Slope = $m$ = difference in y-axis value/difference in x-axis value

$$m = 6.5-4.5/(0.085-0.080)$$

$$= 400$$

Tensile modulus = $400 / (12.30 \times 3.5)$

$$= 9.29 \text{MPa}$$

3.4.2 Flexural Test (3-PT Bending Test)

Flexural strength, also known as modulus of rupture, bend strength, or fracture strength a mechanical parameter for brittle material, is defined as a material's ability to resist deformation under load. The transverse bending test is most frequently employed, in which a rod specimen having either a circular or rectangular cross-section is bent until fracture occurs, using a Three point
bending technique. The flexural strength represents the highest stress experienced within the material at its moment of rupture. It is measured in terms of stress; here given the symbol $\sigma$. Figure 3.8 (a-c) shows the detailed view of the specimens during flexural testing (a) and the specimens (b) and (c).

(a) Specimen placed in flexural testing machine

(b) Specimen before testing
(c) Specimen after testing

Figure 3.8 Flexural testing calculation for flexural modulus and strength
E = FL\(^3/48yI\) (N/mm\(^2\))

where

F is Load in N
L is Length in mm
y is Deflection in mm
E is Youngs modulus in N/mm\(^2\)
I is Moment of inertia
B is Breadth in mm
H is Height in mm

where \(I = BH^3/12 = 17.50 \times 8.60^3/12\)

\(I = 927.58 \text{ mm}^4\)

\(Y = H/2 = 8.60/2 = 4.3\text{mm}\)

Gauge length = \(L = 140\text{mm}\)

Force = 490N

\(E = FL^3/48yI\)

\(E = 490 \times 140^3/(48 \times 1.77 \times 927.58) = 17061.385\text{N/mm}^2\)

\(EI = FL^3/48y\)

\(EI = 490 \times 140^3/(48 \times 1.77) = 15.82 \times 10^6\text{N-mm}^2\)

Stiffness = Force/Deflection = \(460/1.77 = 259.8\text{ N/mm}\)

### 3.4.3 Impact Properties

The measurement of impact strength is an essential part of any material evaluation programme. Most of the test methods are basically simple, but the results, which emerge, are far from straightforward, primarily
because impact strength is not a single, inherent, physical property but rather a combination of several.

Data on impact strength suffer from two deficiencies; the first is that there can be wide discrepancies between results for nominally identical batches and the second is that laboratory results often correlate poorly with service performance. These deficiencies are attributable in part to the well-known sensitivity of impact strength to processing variables, but the second one is attributable also to factors associated with mould and gate geometry in the case of injection mouldings, to the design of extrusion heads, and to the size of the object under consideration.

Izod impact strength testing is an ASTM standard method (ASTM-D256), of determining impact strength. A notched sample is generally used to determine impact strength. Impact is a very important phenomenon in governing the life of a structure. In the case of aircraft, impact can take place by the bird hitting the plane while it is cruising, during takeoff and landing; there is impact by the debris present on the runway. The impact properties of the polymeric materials depend mainly on the toughness of the material.

Toughness can be described as the ability of the polymer to absorb applied energy. The molecular flexibility has a great significance in determining the relative brittleness of the material. Impact energy is a measure of toughness, and the impact resistance is the ability of a material to resist breaking (fracture) under a shock loading. An arm held at a specific height (constant potential energy) is released. The arm hits the sample and breaks it. From the energy absorbed by the sample, its impact strength is determined. Impact strength is so sensitive to molecular orientation, crystalline and crystal texture that some variations in the strength point to
point are to be expected in injection mouldings. Figure 3.9(a-c) shows the impact testing machine and the tested specimens.

(a) Impact testing machine

(b) Before testing(b) After testing

Figure 3.9 Impact test

3.4.4 Durometer Hardness Test (ASTM D 2240)

Hardness is defined as the resistance of a material to deformation, particularly permanent deformation, indentation, or scratching. Hardness is purely a relative term and should not be confused with wear and abrasion
resistance of plastic materials. Since plastic materials vary considerably with respect to hardness, one type of hardness test is not applicable to cover the entire range of hardness properties encountered. Two of the most commonly used hardness tests for plastics are the Rockwell hardness test and the Durometer hardness test. Rockwell hardness test is used for relatively hard plastics such as acetyls, nylons, acrylics, and polystyrene. For softer materials such as flexible PVC, thermoplastic, rubber and polyethylene, durometer hardness test is often used.

The Durometer hardness test is mostly used for measuring the relative hardness of soft materials as has just been stated. The test method is based on the penetration of a specified indentor forced into the material under specified conditions.

The Durometer hardness tester consists of a pressure foot, an indentor, and an indicating device. The indentor is spring loaded and the point of the indentor protrudes through the hole in the base. The test specimens are at least ¼ in.-thick and can be either molded or cut from a sheet. Several thin specimens may be piled to form a ¼ - in.-thick specimen but one piece specimens are preferred. The poor contact between the thin specimens may cause results to vary considerably.

The test is carried out by first placing a specimen on a hard, flat surface. The pressure foot of the instrument is pressed onto the specimen, making sure that it is parallel to the surface of the specimen. The durometer hardness is read within 1 sec after the pressure foot is in firm contact with the specimen is shown in Figure 3.10.
3.4.5 Dynamic Mechanical Analysis Test

Dynamic Mechanical Analysis (DMA), otherwise known as DMA, is a technique where a small deformation is applied to a sample in a cyclic manner. This allows the materials to respond the stress, temperature, frequency and other values to be studied. The term is also used to refer to the analyzer that performs the test. DMA is also called DMTA (Dynamic mechanical thermal analysis). The sample is clamped into a frame of measurement head and is heated by the furnace as shown in Figure 3.11 (a-b). The sample in the furnace is applied the stress from the force generator via probe. To make the strain amplitude constant, the stress is applied in the form of a sinusoidal force.

Figure 3.10 Shore-D- hardness testing apparatus
DMA measures stiffness and damping and these are reported as modulus and tan delta. The storage modulus, either E’ or G’, is the measure of the sample’s elastic behavior. The ratio of the loss to the storage is the tan delta and is often called damping. It is a measure of the energy dissipation of a material.

(a) DMA Apparatus (b) DMA specimen

Figure 3.11 DMA test

3.5 TESTING OF SPECIMENS FOR THERMAL PROPERTIES TEST

The Application of the Thermoplastics in automobile is normally for interior and exterior parts of the automobiles having chances to face various temperature changes. Hence it is important to have evaluation of these properties while designing the materials for automobiles. In Heat Deflection Temperature (HDT), the temperature is being continuously increased where materials can deflect by 0.25mm at an applied force, when the specimens are placed in three point bending mode. The testing of the specimens is taken according to ASTM D 648 with load of 1.08 Mpa. Then heat aging of the
specimens is monitored. Aging of the tensile test specimens is kept in an air oven 120°C for 1000 hrs; as per ASTM standard D3045, testing is performed after 24 hrs. Thermal Gravimetric (TG/DTA) test is also conducted as per ASTM standard. Results are compared with LFRT Specimens

3.5.1 Heat Deflection Test

Heat Deflection temperature is defined as the temperature at which a standard test bar deflects a specified distance under a load. It is used to determine short-term heat resistance. It distinguishes between materials that are able to sustain light loads at high temperatures and those that lose rigidity over a narrow temperature range. This temperature is also known as heat deflection test or Heat Distortion Test (HDT).

The bars are placed under the deflection measuring device. A load of 0.45 MPa or 1.80 MPa is placed on each specimen. The specimens are then lowered into a silicone oil bath where the temperature is raised at 2° C per minute until they deflect 0.25 mm for ASTM, 0.32 mm for ISO flatwise.

Limitations that are associated with the determination of the HDT are that the sample is not thermally isotropic and thick samples in particular, will contain a temperature gradient. The HDT of a particular material can also be very sensitive to stress experienced by the component which is dependent on the component’s dimensions. The selected deflection of 0.25 mm (which is 0.2% additional strain) is done arbitrarily and has no physical Meaning. Figure 3.12(a-b) shows the HDT apparatus and specimen.
(a) HDT apparatus (b) Tested specimen

Figure 3.12 HDT testing

ASTM uses a standard bar 5" x ½" x ¼". ISO edgewise testing uses a bar 120mm x 10mm x 4mm. ISO flatwise testing uses a bar 80mm x 10mm x 4mm.

The load (F) applied to the sample will vary with thickness (t) and width (w) of the samples and is determined by the maximum stress specified at the mid-point of the beam (p) which may be either 45MPa or 1.82 MPa.

The formula used for the calculation is:

Model calculation for HDT

\[ F = \frac{2Pw^2}{3L} \]

F = Load in N

P = deflection in mm

w = width in mm
deflection \( P = 1.8 \text{N/mm}^2 \) (constant)

\[
F = \frac{2 \times 1.8 \times 13 \times 3.5^2}{3 \times 127} = 0.275866 \text{ N}
\]

By Newton’s second law,

\[
F = m \times a
\]

\[
m = \frac{F}{a} = 0.2758/9.81 = 0.0281 \approx 2.8 \text{ gms}
\]

Heat deflection temperature for sample A at load of 2.8 gms is 56 °C.

### 3.5.2 TG-DTA Test

Thermo gravimetric analysis or Thermal Gravimetric Analysis (TGA) is a method of thermal analysis in which changes in physical and chemical properties of materials are measured as a function of increasing temperature (with constant heating rate), or as a function of time (with constant temperature and/or constant mass loss).

TGA is commonly used to determine selected characteristics of materials that exhibit either mass loss or gain due to decomposition, oxidation, or loss of volatiles (such as moisture).

Thermo gravimetric Analysis is a technique in which the mass of a substance is monitored as a function of temperature or time as the sample specimen is subjected to a controlled temperature program in a controlled atmosphere. An alternate Definition: TGA is a technique in which, upon heating a material, its weight increases or decreases. A Simple TGA Concept
to remember: TGA measures a sample’s weight as it is heated or cooled in a furnace.

The balance beams for the sample and the reference are located in the furnace. The masses of the sample and the reference are measured by the sensitivity-calibrated drive coils separately. The mass difference is sent as TG signal. By the differential mass measurement, the effects of the beam expansion, the convection flow, and buoyant force are cancelled. Thus the highly sensitive thermo gravimetric measurement is achieved. The mass measurement of the sample and the reference by the independent drive coils enables the easy adjustment of the TG baseline drift electrically. Also, thermocouple is located in each holder which enables the simultaneous DTA signal output. TG can be utilized for the analysis of the thermal decomposition, the oxidization, the dehydration, the heat resistance, and kinetics analysis. By combining with the other measurement technique, variety of information can be achieved from one sample. In particular, TG/DTA simultaneous measurement instrument is most common.

Thermo gravimetric analysis or TGA is a type of testing performed on samples that determines changes in weight in relation to change in temperature. Such analysis relies on a high degree of precision in three measurements: weight, temperature, and temperature change. As many weight loss curves look similar, the weight loss curve may require transformation before results may be interpreted. To determine composition and purity one must take the mass of the substance in the mixture by using thermal gravimetric analysis. It is a process that utilizes heat and stoichiometry ratios to determine the percent by mass ratio of a solute. If the compounds in the mixture that remain are known, then the percentage by mass can be determined by taking the weight of what is left in the mixture and dividing it by the initial mass. TGA is commonly employed in research
and testing to determine the characteristics of materials such as polymers, to determine degradation temperatures, absorbed moisture content of materials, the level of inorganic and organic components in materials, decomposition points of explosives, and solvent residues. It is also often used to estimate the corrosion kinetics in high temperature oxidation.

![Figure 3.13 TG-DTA apparatus](image)

**Figure 3.13 TG-DTA apparatus**

TGA is a process that utilizes heat and stoichiometry ratios to determine the percent by mass of a solute. Analysis is carried out by raising the temperature of the sample gradually and plotting weight (percentage) against temperature. The temperature in many testing methods routinely reaches 1000°C or greater. After the data are obtained, curve smoothing and other operations may be done to find the exact points of inflection. Figure 3.13 shows the TGA/DTA apparatus.

### 3.5.3 Heat Aging

The heat aging analysis is important to monitor the aging capacity of the natural fibers during the temperature deviations for long durations, since the usage is for the automotive applications where the aging of the materials
deals with the quality of the vehicle. In the present study, the heat aging of the specimens was monitored. Aging of the tensile test the specimens were kept in an air oven 120ºC for 1000 hrs, as per ASTM standard D3045 and testing was performed after 24 hrs. It is shown in Figure 3.14.

![Figure 3.14 Heat aging tested specimens](image)

**Figure 3.14 Heat aging tested specimens**

### 3.6 RECYCLING OF COMPOSITES

Kenaf fibers are less brittle and softer than glass fibers and are likely to result in composites that are easier to recycle than mineral based fibers. Thermoplastics have more recyclability compared to thermosetting plastics. Grinding reprocessing is general method to recycle the thermoplastics. This recycling process is efficient and economic compared to other recycling methods like chemical recycling, particle recycling and energy recycling. To analyze the recycling property, KLFRT, JLFRT, SLFRT and LFRT materials were reground followed by injection moulding process. The regrinding and injection molding process was repeated twice for better comparison of properties like tensile properties. The recycled tensile properties values were compared with virgin values of the composite.
The testing of the mechanical, thermal and recycling of the composite is performed as per the ASTM. The correct test speed, loading profile and methodology for calculations were followed as per the standards. The rate at which a test is performed can have a significant effect on testing results. The above mentioned tests were carried out in well calibrated instruments which are having good accuracy and working conditions. There are six to eight sets of the readings were taken with standard testing rate and specified range to ensure the accurate tested values.

3.7 SUMMARY

This chapter describes the details of the processing of the composites using hot impregnation process using natural fibers like kenaf, sisal and jute. The matrix composition and the fiber loading percentage are decided and it is very important to decide the strength of the composites. By using the hot pultrusion process the long fiber pellets are prepared. The specimens are created for the KLFRT, SLFRT, JLFRT and LFRT composites and are injection moulded from pellets according to ASTM standards and the experimental testing procedures followed for evaluate the mechanical, thermal and recycling characterization.