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Natural Fibers/Fillers Reinforced Composite: A Review

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ABSTRACT: *In recent era, bio-fillers have increased their demand due to several advantages like low expenditure, low weight, low density, biodegradability, non-toxicity, renewable, easy availability, low health hazards and prevention from deadly chemicals and environmental attacks. However, their poor interfacial adhesion with some matrix material leads to poor/low mechanical properties of the composites. This review presents study of various natural fibers/fillers such as coir, flax, jute, nutshell, walnut shell, groundnut shell, coconut shell etc. which can be used in various industries when mixed with certain reinforcement results in improving mechanical properties such as tensile strength, flexural strength, hardness and impact properties. These natural reinforced composite have large numbers of applications in various industries ranging from defense industry, furniture, automotive industry, space industry and many more. The aim of this paper is to study the mechanical properties of different natural fibers reinforced polyester composites. The specimens were prepared in to the polyester matrix. The particles of wt. % 0 to 40 were taken in the experimental study. The results of prepared composites will then be compared with neat polymer specimen.*

KEYWORDS: Natural Fibers/Fillers, Reinforced Composite, tensile strength, flexural, hardness, impact factor.

INTRODUCTION

Alarming environmental scenario in the present century motivates the researchers worldwide on the studies of natural fiber reinforced polymer composite and cost effective option to synthetic fiber reinforced composites. Increased demand for raw materials in various industries as a result of population growth and depletion of the natural resources, have directed researchers towards bio-based alternatives for composite manufacturing. Agricultural residues are excellent alternative materials to wood because they are inexpensive, easily processed, plentiful, and renewable [1,2].Some of the problems associated with industrial usage of agricultural residues the forest industry include the high cost of collection, transportation and storage that can be overcome by establishing small-scale plants close to rural areas [3].

Fiber-reinforced composites material offer high strength to weight ratio, improved dimensional stability, better environmental resistance and degradability as compared to conventional composites. In these days, polymer composite materials are extensively used in engineering applications due to their excellent specific physical and chemical properties. Natural fiber reinforced composites applications are in automotive, industrial, building, and other commercial markets. Various parts of automobiles like door panels, headliners, package trays, dashboards and interior parts are being produced using these natural fiber based composites [4]. Natural fiber reinforcements in the form of short fiber, filament or fabric have become better alternatives to synthetic fibers as reinforcements,

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due to their high flexural modulus and strength as well as impact strength and modulus. They are less costly, lighter in weight, lower in density and higher in specific strength, non-corrosive and easier to manufacture [5-7]. The biodegradability of natural fibers is well in line with the demands of a healthier environment, while their high performance and affordable cost satisfies the economic benefit of industries [8].

Walnut shell, coconut shell, Kans grass, Pineapple leaf, oil palm fiber, hemp, sisal, Jute, kapok, rice husk, bamboo, coir, flax, ramie, sisal, jute, banana, luffa cylindrica, bagasse are some examples of natural fibers which are most commonly used as reinforcing materials in polymer composite industry [8]. These days, polypropylene (PP), polyethylene, and poly vinyl chloride dominate as matrices for natural fibers. Polypropylene (PP) is extensively used as advanced composite matrix because it is cheap, strong and easily process able, while polypropylene-based lining material is now in use owing to its superiority in resistance to wear [9].

Composites can be defined as the combination of a matrix and reinforcement, which when combined together gives property that is superior to the property of the individual components [10]. The reinforcement fibres can be cut, aligned and placed in different ways to affect the properties of the resulting composite. The matrix, normally a form of resin, keeps the reinforcement in the desired orientation. It protects the reinforcement from chemical and environmental attack, and it bonds the reinforcement so that applied loads can be effectively transferred [11]. Many composite materials are composed of two phases; one is termed as matrix, which is continuous and surrounds the other phase, often called the dispersed phase. "Dispersed phase geometry" means the shape of the particles and the particle size, distribution, and orientation [12].

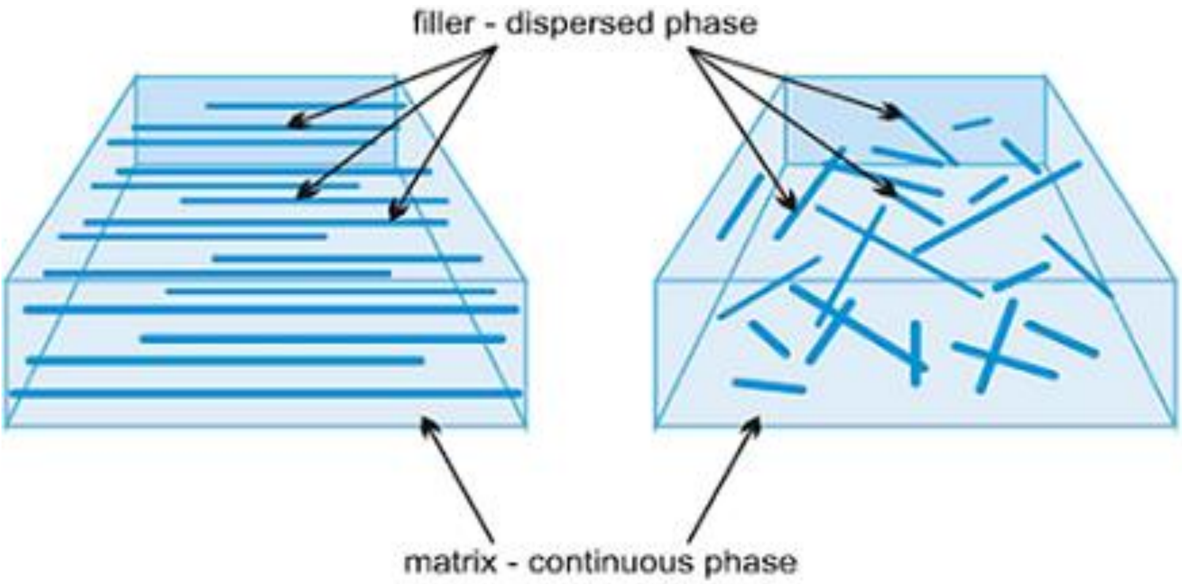

Figure1: Matrix and dispersed phase of composites

Classification of composites

Composite materials can be classified in two different ways. Figure 2 and 3 illustrate the classification clearly.

1. On the basis of geometry of a representative unit of reinforcement.
2. On the basis of matrix material.

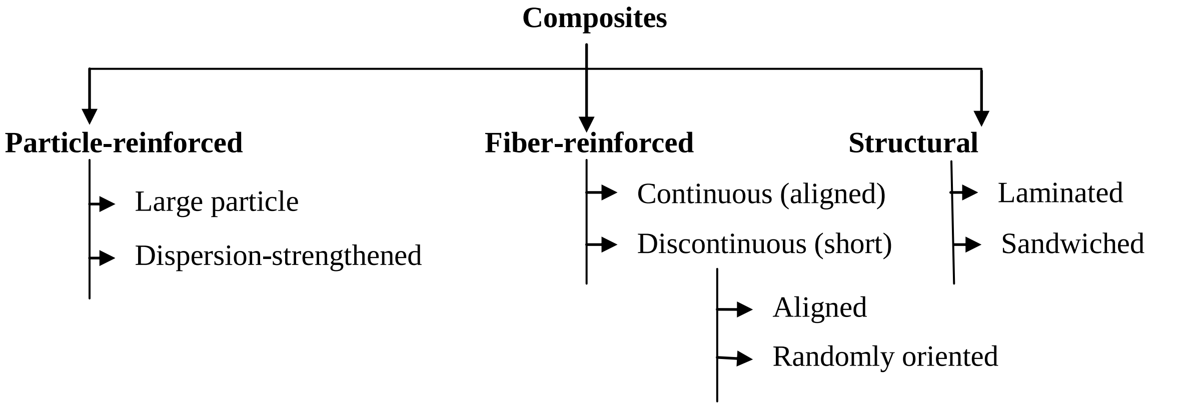

Figure 2: On the basis of reinforcement

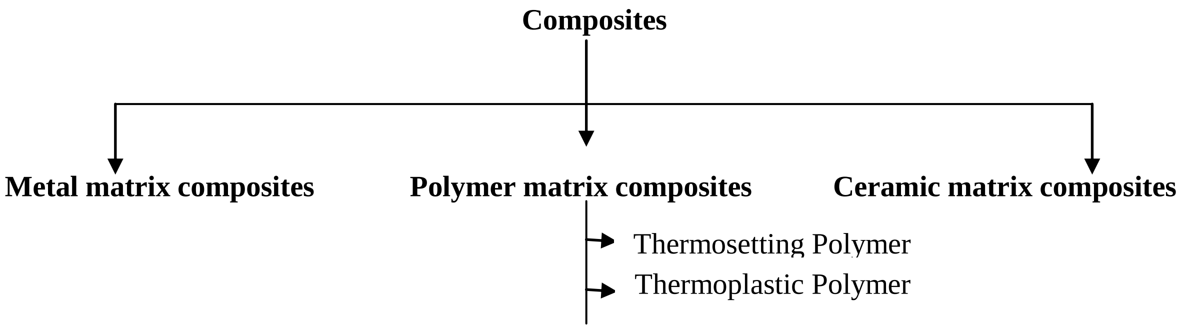

Figure 3: On the basis of matrix material composites

On the basis of geometry of a representative unit of reinforcement

a) Particle-reinforced composites

As its name indicates, the reinforcement is of particle nature. It may be spherical, cubic, tetragonal, a platelet, or of other regular or irregular shape. In general, particles are not very effective in improving fracture resistance but they enhance the stiffness of the composite to a limited extent. Particle fillers are widely used to improve the properties of matrix materials such as to modify the thermal and electrical conductivities, improve performance at elevated temperatures, reduce friction, increase wear and abrasion resistance, improve mach inability, increase surface hardness and reduce shrinkage. Particles are used to increase the modules of the matrix and decrease the ductility of the matrix [13].

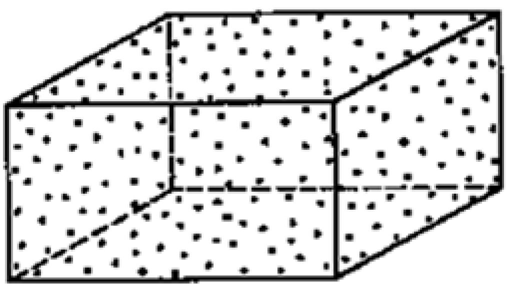

Figure 4: Particles reinforced composite

Polymer composite materials have generated wide interest in various engineering fields, particularly in aerospace applications. Research is underway worldwide to develop newer composites with varied combinations of fibers and fillers to make them usable under different operational conditions.

These composites are the cheapest and most widely used. They fall in two categories depending on the size of the particles:

- Large-particle composites, which act by restraining the movement of the matrix, if well bonded.
- Dispersion-strengthened composites, containing 10-100 nm particles, similar to what was discussed under precipitation hardening. The matrix bears the major portion of the applied load and the small particles hinder dislocation motion, limiting plastic deformation.

b) Fiber reinforced composites

Dispersed phase in form of fibers (Fibrous Composites) improves strength, stiffness and fracture toughness of the material, impeding crack growth in the directions normal to the fiber. Increase in strength becomes much more significant when the fibers are arranged in a particular direction and a stress is applied along the same direction. The strengthening effect is higher in long-fiber (continuous-fiber) reinforced composites than in short-fiber (discontinuous-fiber) reinforced composites [14].

Fiber reinforced composites are again classified into two groups based on the length of fibers used. They are as follows:

- Continuous Fiber reinforced composites
- Discontinuous Fiber reinforced composites

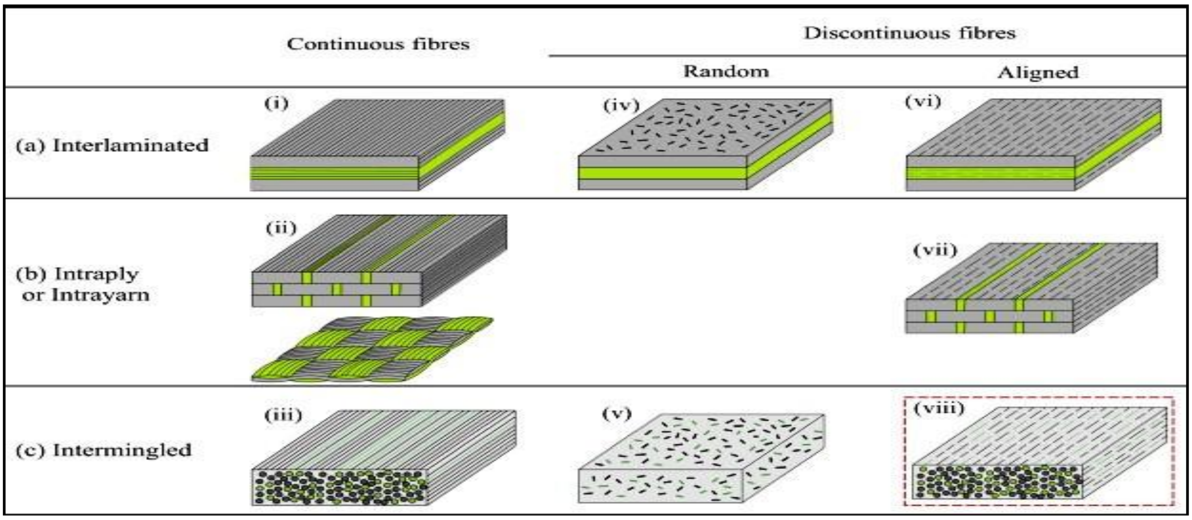

Figure 5: Continuous and discontinuous fiber composites

The discontinuous fiber composites are further classified as: aligned FRC's and random FRC's (based on orientation of fibers). The interest in the natural fiber reinforced polymer composites is growing rapidly due to the following benefits: High Specific strength, Easy availability, Light weight, Ease of separation, Non-corrosive nature, Low density, Cost effective, Reduced tool wear, Less abrasion to processing equipment, Renewability and biodegradability.

c) Structural composites

These types of composites can be classified into two types. One is laminated type and the second is sandwich type. Laminate composites consist of different layers of matrix reinforced with a dispersed phase in form of sheets. When a fiber reinforced composite consists of several layers with different fiber orientations, it is called multilayer (angle-ply) composite. Laminate composites provide increased mechanical strength in two directions and only in one direction, perpendicular to the preferred orientations of the fibers or sheet. Sandwich-structured composite is a special class of composite materials that is fabricated by attaching two thin but stiff skins to a lightweight but thick core. The core material is normally low strength material, but its higher thickness provides the sandwich composite with high bending stiffness with overall low density.

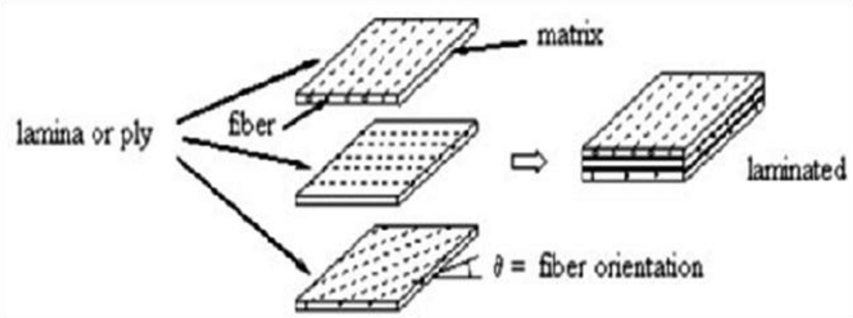

Figure 6: Laminated (Multilayer) fibers composites

On the basis of matrix material composites

a) Metal Matrix Composites

Metal matrix composites (MMCs) are composite materials that contain at least two constituent parts – a metal and another material or a different metal. The metal matrix is reinforced with the other material to improve strength and wear. Where three or more constituent parts are present, it is called a hybrid composite. Examples of matrices in such composites include aluminum, magnesium and titanium. The typical fiber includes carbon and silicon carbide. These types of composites metal matrix composites have many advantages. MMCs are fire resistant, operate in a wide range of temperatures, do not absorb moisture, and possess better electrical and thermal conductivity. They have also found applications to be resistant to radiation damage, and to not suffer from out gassing. Most metals and alloys make good matrices for composite application. Because of these attributes metal matrix composites are under consideration for wide range of applications viz. combustion chamber nozzle (in rocket, space shuttle), housings, tubing, cables, heat exchangers, structural members etc.

b) Ceramic Matrix Composites

Ceramic matrix composites (CMCs) are a subgroup of composite materials. They consist of ceramic fibers embedded in a ceramic matrix, thus forming a ceramic fiber reinforced ceramic (CFRC) material. The matrix and fibers can consist of any ceramic material. CMC materials were designed to overcome the major disadvantages such as low fracture toughness, atmospheric and other forms of corrosion, and exhibit superior resistance to the conduction of electrical current. Ceramic matrix composites have ceramic matrix such as alumina, calcium, alumina silicate reinforced by silicon carbide. The advantages of CMC include high strength, hardness, high service temperature limits for ceramics, chemical inertness, and low density [15].

c) Polymer Matrix Composites

Polymer matrix composites (PMCs) can be divided into three sub-types, namely, thermoset, thermoplastic, and rubber. Polymer is a large molecule composed of repeating structural units connected by covalent chemical bonds. PMC's consist of a polymer matrix combined with a fibrous reinforcing dispersed phase. PMC's are less dense than metals or ceramics, can resist atmospheric and other forms of corrosion, and exhibit superior resistance to the conduction of electrical current. These materials can be fashioned into a variety of shapes and sizes. They provide great strength and stiffness along with resistance to corrosion. The reason for these being most common is their low cost, high strength and simple manufacturing principles. Due to the low density of the constituents, the polymer composites often show excellent specific properties. The processing of polymer matrix composites need not involve high pressure and do not require high temperature. Also, equipment's required for manufacturing polymer matrix composites are simple. For this reason, polymer matrix composites developed rapidly and became popular for structural applications [15]. Now days polymers like polyester, epoxy resins etc. are most widely used for composites development due to their favorable properties.

Polyester

Polyester is the type of thermosetting polymers. Thermosets are solidified by irreversible chemical reactions in which the molecules in the polymer “cross-link”, or form connected chains. Polyester is an extremely versatile, fairly inexpensive polymer. Unsaturated polyester combines an unsaturated dibasic acid and a glycol dissolved in a monomer, generally styrene, including an inhibitor to stabilize the resin. Polyester resins are less expensive, offer some corrosion resistance, and show good performance. The majority of all fiber parts are constructed using polyester resins because they are easy to use, fast curing, and tolerant of temperature and catalyst extremes [16].Table 1 shows some typical properties of thermoset resins.

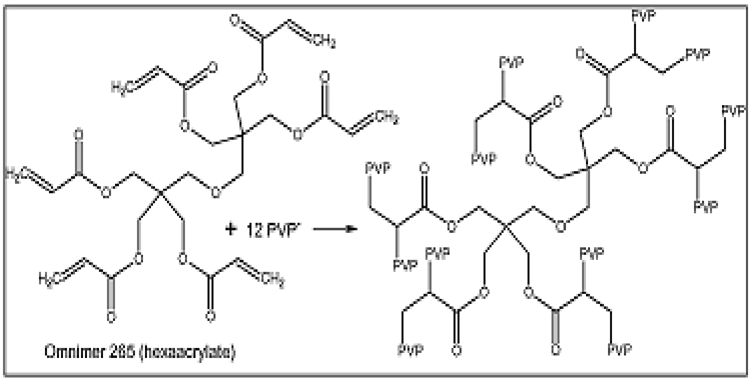

Figure 7: Cross-linked chain of polyester

Table 1 Typical properties of different thermosets

Properties	**Epoxies**	**Polyesters**	**Phenolics**	**Polyimides**
Density, gm/cm^3	1.1-1.4	1.1-1.3	1.2-1.3	1.3- 1.4
Tensile strength, MPa	35-100	50-60	50-60	100-130
Tensile modulus, GPa	1.5-3.5	2-3	5-11	3-4
Poissons ratio	0.35	0.35	0.35	0.35
Coefficient of thermal expansion, 10^{-6}m/mK	50-70	40-60	40-80	30-40
Service temperature, K	300-370	330-350	440-470	550-750

Natural Fibers

Natural fibers include those produced by plants, animals and geological processes. The majority of useful natural fibers are plant derived, with the exceptions of wool and silk. All plant fibers are composed of cellulose, whereas fibers of animal origin consist of proteins. Natural fibers in general can be classified on the basis of their origin of the plant. Some of the examples are:

- **Bast Fibers:** Flax, Hemp, Jute, Kenaf, Ramie, Banana
- **Leaf Fibers:** Sisal, Manila, Banana, Palm

- **Seed Fibers:** Cotton, Kapok
- **Fruit Fibers:** Coconut, coir, walnut
- **Straw Fibers:** Wheat, Rice, Barley
- **Wood Fibers:** Hardwood, Softwood, Walnut, Argon nut shell
- **Grass Fibers:** Kans grass, Sorghum, Bamboo

Table 2: shows the strength of various natural composite [17]

Properties	Fibers								
	E-glass	Flax	Hemp	jute	ramie	Coir	Sisal	abaca	Cotton
Density g/cm^3	2.55	1.4	1.48	1.46	1.5	1.25	1.33	1.5	1.51
Tensile strength$^*10^6$N/m^2	2400	800-1500	550-900	400-800	500	220	600-700	980	400
E-modulus (GPa)	73	60-80	70	10-30	44	6	38	-	12
Specific (E/density)	29	26-46	47	7-21	29	5	29	-	8
Elongation at failure (%)	3	1.2-1.6	1.6	1.8	2	15-25	2-3	-	3-10
Moisture absorption (%)	-	7	8	12	12-17	10	11	-	8-25
Price ($),raw(mat/fabric)	1.3 (1.7/3.8)	1.5 (2/4)	0.6-1.8 (2/4)	0.35 1.5/0.9-2	1.5-2.5	0.25-0.5	0.6-0.7	1.5-2.5	1.5-2.2

Method of Composite Fabrication

The fabrication of composite involves the combination of the matrix and fiber or reinforced particles such that the matrix impregnates surrounds and wets the fibers. The process can be adopted on the basis of matrix materials used like thermosetting or thermoplastic materials. The important processing methods used for thermosetting polymer are Hand layup, Bag molding process, Filament winding, Pultrusion, Bulk molding, Sheet molding, Resin transfer molding and Scrap-rotate-scrap mixing.

Avila A.*et al*. [18] worked on fiber glass-epoxy-nano-clay laminate composites to investigate the plate impact strength in the presence of nano-clay. They prepared S2- glass/epoxy-nano-clay nano-composites for this. The S2-glass/epoxy-nano-clay composite is a laminate with 16 layers and 65% fiber volume fraction. This type of configuration is prepared using a vacuum assisted lay-up technique. The amount of nano-clay added into an epoxy system in weight1%, 2%, 5%, 10%, respectively. To compare the results a set of S2-glass/epoxy laminated composites is also prepared. The addition of nano-sized clays increases the composite impact strength, as the damaged area is decreased approximately 20% for small amounts of nano-clay contents. When the concentration reaches around 10% the increase on impact strength is near to 50%. Nano-clay composites show great performance over the conventional composites under the rebound/spring effect.

The comparative study developed by C**. C. Ugoamadi *et al.* [19]**on cashew nuts picked and processed for the extraction of the resin content was mixed with cobalt amine (accelerator), methyl ethyl ketone peroxide (catalyst) to develop two sets of composite specimens – specimens without fibres and specimens reinforced with glass fibres. This method of sample specimen development was repeated with polyester (synthetic) resin. Compressive and tensile strength tests conducted proved that composites developed with cashew nut shell liquid (CNSL) resin were comparable to those developed with polyester resin. In the results, CNSL has an ultimate compressive strength of 55MPa compared to that of polyester resin with an ultimate strength of 68MPa. The result of tensile strength proved cashew nut shell liquid resin (with ultimate strength of 44MPa) to be better than polyester resin with 39MPa as ultimate tensile strength. This means that natural resins could be a better substitute for the synthetic ones when the required quantities of fibers (reinforcements) and fillers are used in the fibre-reinforced plastic composite developments.

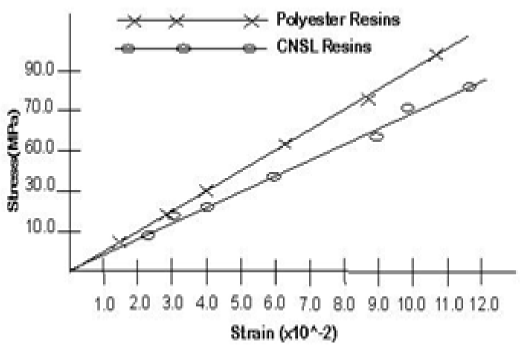

Figure 8: Graph of compressive stress against strain of unreinforced polyester and CNSL resins

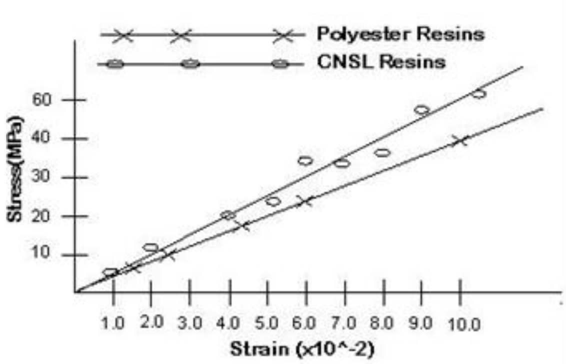

Figure 9: Graph of tensile stress against strain of unreinforced polyester and CNSL resins

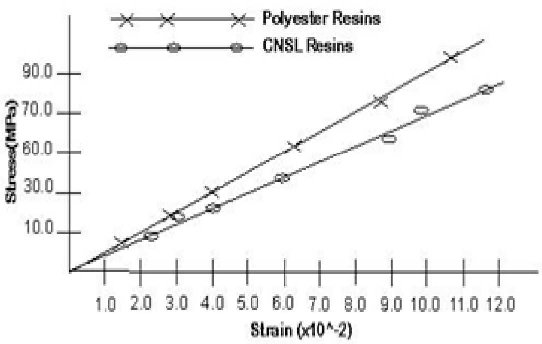

Figure 10: Graph of compressive stress against strain of polyester and CNSL resins reinforced with glass fibres

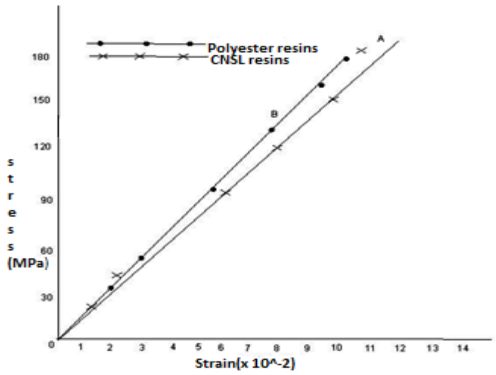

Figure 11: Graph of tensile strength against strain of polyster and CNSL resins reinforced with glass fibers

Figure 8 and 10 shows the same results because the inclusion of fibers in polymer matrices does not change the compressive strength of the resulting composite. In figure 11 there is remarkable increase in the tensile strengths of the composites developed from the two polymer matrices as fibre reinforced composite materials are subjected to tensile loading, the fibre absorb the major component of the load and this makes the fibre to be responsible for the tensile strength of any composite.

Chensong Dong *et al.*[20] investigated flexural properties of macadamia nutshell particle reinforced polyester composites. Four weight fractions of nutshell particles 10%, 20%, 30% and 40% were chosen to be studied. When voids exist, flexural modulus was calculated using a micromechanical model. The experimental data shows that the flexural modulus increases with the weight fraction of macadamia nutshell particles, while decreases with increasing void content. Adding macadamia nutshell particles does not improve the flexural strength of polyester. Thus, flexural strength decreases with increasing void content.

Kamila salasinka*et al*. [21] aimed to develop composites of waste materials from sunflower husk and pistachio shell without the use of additives. It was found that 66% of the sunflower husk grains were from 180 to 850 μm in size, and 88% of the pistachio shell particles were less than 63 μm in size. With the use of a rolling mixer, six mixtures were produced with filler shares amounting to 5, 15 and 30 wt.% of sunflower husk and 15, 35 and 55% of pistachio shell, from which dumbbell-shaped samples were formed via injection processing. The produced materials were analyzed for their mechanical properties (impact strength, hardness, tensile strength, Young's modulus and DMA). The produced composites were considered to be solid materials, with porosities below 5 %. The NFC porosities increased with increasing filler content, and voids occurred primarily in the contact regions of the composites and were the result of their poor adhesion.

Beckry *et al.* [22] examined the properties of an E-glass/epoxy composite before and after mechanical loading and moisture conditioning. Preliminary results indicate that the modulus, strength, and strain of the E-glass/epoxy composite material are affected by the presence of moisture and mechanical loading when compared to control specimens. At shorter durations of conditioning at room temperature, a slight increase in strength and a slight decrease in modulus were observed and at longer duration i.e.3000 h, a noticeable reduction in strength and strain-to-failure was observed. Specimens conditioned under stress, in water at 650˚C for 1000 h exhibited higher loss in modulus. It is speculated that constant stress may have a positive effect in short-term, and that extended exposure to moisture at room temperature leads to brittle failure while exposure at high temperatures may lead to ductile failure of E-glass/epoxy composites.

Rao D.K. *et al*[23] prepared 20 wt % of walnut shell particles and 10 wt % of coconut fibres were used as a reinforcing material in epoxy resin CY 230 and hardener HY 951. Hardener was mixed in the solution at 400˚ C which were preheated to 1000˚ C and hold for 2 hours at 1000˚ C. After curing, the composite sheet was used for tensile test to fulfill the objectives of the present investigation. A simple specimen shape according to ASTM D 790 is used for the flexural test.

The thickness and width of the specimen are measured and recorded. This test is conducted using servo hydraulic universal testing machine (ADMET). The force displacement curve for bio-composite (20 wt % walnut shell particle and 10 wt % coconut fibres) and pure epoxy material. The curve rises in non linear way until the maximum value reached where load drop suddenly. The non linear increase in the load demonstrates the ductile behavior of the material and this may be because of the use of short coconut fibre. The average values of ultimate strength of biocomposite and pure epoxy are 24.43 MPa and 44.93 MPa respectively. This shows that the ultimate strength of biocomposite is about 54.3% of pure epoxy. However the coefficient variance of biocomposite as compared to pure epoxy is about 6 times of the pure epoxy.

Meysam Zahedi*et al*. [24] studied the characterization of walnut shell/PP composites and compared with wood flour/PP composites. The effects of organo-montmorillonite (OMMT, 0, 3 and 5 wt. %) as reinforcing agent and MAPP as coupling agent (4 and 6 wt. %) on the mechanical and physical properties were also investigated. Composites manufactured via melt compounding and subsequent hot press. The resulting composites had acceptable and comparable properties to wood flour properties and the highest improvement of mechanical properties was achieved at 3% organo-clay loading and 6% MAPP. Morphologies of the nano-composites were analyzed by scanning electron microscopy (SEM) and transmission electron microscopy (TEM). SEM study approved the good interaction of the walnut shell flour with the polymer as well as effectiveness of organo-clay in improvement of the interaction. TEM study revealed better dispersion of silicate layers in polymer–matrix composites (PMCs) loaded with 3 wt. % of clay.

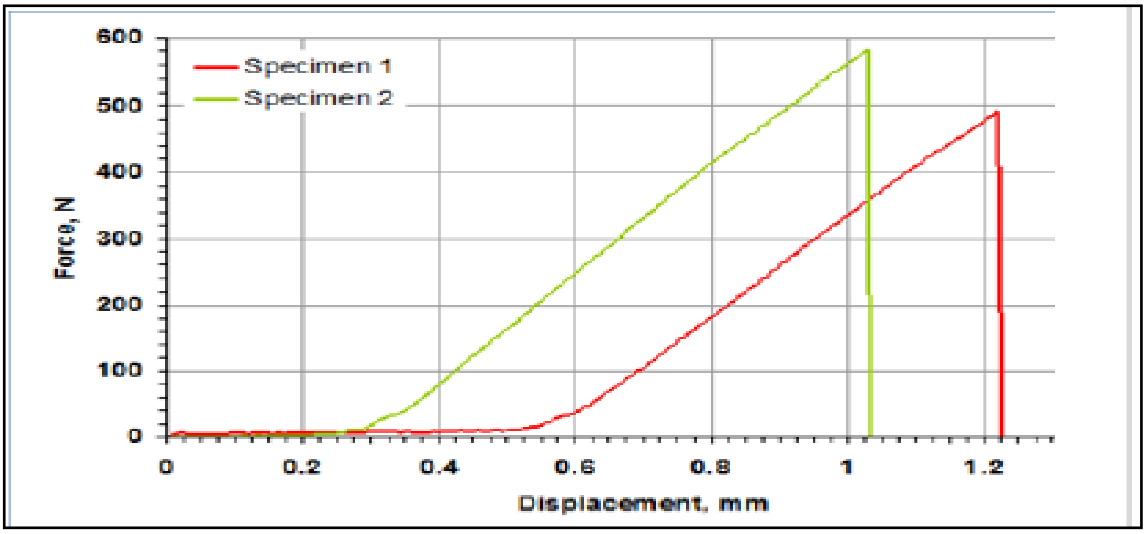

Figure 12: Shows the force displacement curve for bicomposite with 20% walnut shell particle and 10% coconut fibre [23].

M.Guru, *et al*. [25]developed walnut shell urea–formaldehyde based composite particle board from walnut shell and improved its requirements such as non-flammability, eradication of fungal and insect attack, and water resistance features by using fly ash and phenol–formaldehyde. For this purpose, under laboratory conditions, parameters affecting polymer composite particleboard from walnut shell and urea–formaldehyde were specified to be urea–formaldehyde ratio, reaction temperature, and reaction period, and the effect of these parameters on hardness was investigated. The optimization results showed that maximum bending strength was 3.8 N/mm^2, at urea–formaldehyde ratio of 1.0, reaction temperature of 70 °C, reaction time of 25 min, walnut shell filler/urea–formaldehyde resin of 3 and mean particle size of 0.12 mm. As a result of burning tests, although the maximum flame temperature of composite particle board without fly ash was 535 °C, it was 299 °C with fly ash of 15% (w/w) according to the filler. Water absorption and increasing in the thickness exponentially decreased with increasing phenol–formaldehyde/urea–formaldehyde ratio.

Nadir A. *et al* [26] investigated the walnut shell flour and polypropylene based composites with and maleic anhydride-grafted polypropylene at 40, 50 and 60 weight percentage of the walnut shell for mechanical properties. The bending and tensile modulus of the composites significantly increased with increasing the filler content while the bending and tensile strength significantly decreased. Water absorption and thickness swelling of the composites increased with increasing filler content. The MAPP improved the interfacial adhesion between walnut shell flour and polymer matrix. This research revealed that 40/57/3 formulation of the walnut shell flour/ polypropylene/MAPP can be used in outdoor applications a high dimensional stability.

Onat A. *et al* [27] demonstrated that glass fiber based warp knitting and walnut shell reinforcements have impacts on mechanical and thermal properties of composite materials. Tests were applied to composite materials in 0º and 90º directions. Results of all mechanical tests are higher in 0º when compared to the results obtained in 90º direction. This difference of values results from different properties of fiber bundles forming the glass based biaxial fabric used in the production. Difference emerging between 0º and 90º directions as a result of these orientation properties is attributed to the biaxial glass based fabric. According to the results obtained from mechanical tests, a decrease in the particle size of the walnut shell used in the production of composite material yields better results. When the particle size is small, a better interface forms between matrix material and resin and, in turn, higher mechanical values were obtained. When heat conductivity results of composite materials were examined, it is observed that a higher heat conduction coefficient DT is obtained, as the particle gets bigger. It is understood from these results that when bigger walnut shells are used, produced composite materials isolate the heat more effectively.

RESULTS AND DISCUSSION

On the basis of experiment performed by different researchers as quoted above using different natural fibers/fillers reinforced with matrices to form composites with improved mechanical properties (tensile strength, flexural strength, hardness),the following results can be drawn. The percentage of void content increases with increase in the natural fibers/particles up to a certain wt.% and decreases gradually after a certain limit. Increase in void content indicates the bad quality of composites and variation in mechanical properties. The increase in wt. % decreases the tensile strength of composites as compared to neat polyester. The tensile modulus of composite specimen at high wt. % is found to be greater than the neat polyester. The flexural strength of the composite specimens fluctuates as the wt.% increases. The composites hardness increases with increase in wt. % of particles.

CONCLUSIONS

The following conclusions can be made from the literature:

1. Natural Fibres/Fillers from different sources like bast, leaf, seeds, fruits, straw, wood and grasses when combine with different matrices plays an essential components of composites since their presence enhances the mechanical properties such as tensile stress, flexural strength, hardness and impact properties which should be considered as close substitutes for synthetic ones.

2. These natural reinforced composite have large numbers of applications in various industries ranging from defense industry, leather industry, furniture industries, automotive industry, sea transport industry and sport materials which can be produces at low cost, renewable, sustainable, can be moulded to acquire specific strength, low weight, non-toxic to living beings and eco-friendly as well.

3. Non-food crops and other bio-renewable resources offer an almost limitless supply of renewable and potentially sustainable raw materials for the production of biocomposites. Although in its infancy, there is a growing market for biocomposite-based products and with further development a whole host of new applications can be envisioned.

4. There is a large number of potential reinforcing fibres/fillers and an extensive range of processing options to ensure the right fibre at the right cost. In parallel, significant developments have been seen in the horizon of biopolymers in recent years. These combined ensure that biocomposites are likely to see a period of sustained growth through vast research and development which ultimately helps in utilizing renewable resources in place of synthetic toxic substances.

5. Addition to it, use of these renewable resources plays a significant role in providing employment to rural areas and promote the establishment of different small scale industries at very low processing cost with no health hazards thus helps in removing unemployment in developing countries.

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