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Coconut Shell Powder Reinforced Epoxy Composites: A Review

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ABSTRACT

Fibers are of two type's natural fiber and synthetic fiber. Natural fibers include those made from plant and animal sources. The natural fiber composites can be very cost effective material and have turned out to be an alternative solution to the ever depleting petroleum sources and have reduced the nuisance of pollution. The production of complete natural fiber based materials as a substitute for petroleum-based products would not be an economical solution. A more viable solution would be to combine petroleum and bio-based resources to develop a cost-effective product with diverse applications. The application of natural fiber-reinforced composites has been extended to almost all fields such as building and construction industry, storage devices (post-boxes, grain storage silos, bio-gas containers *etc*.), furniture and electrical devices. Coconut shell is a lignocellulosic agro waste which is burnt or left to decay in environment. These can be a very interesting material as filler in biodegradable polymer composites, due to its good thermal stability compared to other agricultural waste. In context to this, review was carried out to assess the coconut shell powder reinforced epoxy composites and their mechanical, structural and thermal properties.

Key words: Biodegradable, Coconut shell powder, Epoxy, Fiber.

Composites are made by combining two or more natural or artificial materials to maximize their useful properties and minimize their weaknesses. Composites generally comprise a strong load carrying material (known as reinforcement) imbedded in weaker material (known as matrix). Reinforcement provides strength and rigidity, helping to support structural load. The matrix or binder (organic or inorganic) binds the fibers together somewhat like an adhesive and makes them more resistant to external damage and maintains the position and orientation of the reinforcement. However both retain their individual physical and chemical properties yet together provide combined effect (Taj *et al.,* 2007). The nature offers many natural composites such as wood consisting of cellulose, hemicellulose and lignin, bone, teeth etc. and from many years our ancestors have developed and used composites. The first uses of composites date back to the 1500 B.C. when early Egyptians and Mesopotamian settlers used a mixture of mud and straw to create strong and durable buildings. Straw continued to provide reinforcement to ancient composite products including pottery and boats. Later, in 1200 AD, the Mongols invented the first composite bow, using a combination of wood, bone and animal glue. The modern era of composites aroused when synthetic polymers were developed. Until then, natural resins derived from plants and animals were the only source of glues and binders (www.thoughtco.com). However plastic alone could not provide enough strength therefore reinforcement was required for additional strength and rigidity. Various fibers such as carbon, boron, silicone and aramids are used as reinforcing material in different matrices like metal, polymer and ceramics with various applications. The greatest advantage of composite materials is strength and stiffness combined with lightness, therefore careful selection of reinforcement type enables finished product characteristics to be tailored to almost any specific engineering requirement (Chanap, 2012).

The composites can be classified into three groups Metal Matrix Composites, Ceramic Matrix Composites and Polymer Matrix Composites. Metal matrix composites, as the name implies, have a metal matrix. Examples of matrices in such composites include aluminum, magnesium and titanium. The typical fiber includes carbon and silicon carbide. Ceramic matrix composites have ceramic matrix such as alumina, calcium, alumino-silicate reinforced by silicon carbide. The most common advanced composites are polymer matrix composites. These composites consist of a polymer thermoplastic or thermosetting reinforced by fiber (natural carbon or boron). These materials can be fashioned into a variety of shapes and sizes. They provide great strength and stiffness along with resistance to corrosion.

However, in recent years there is a constantly increasing demand for biodegradable composites reinforced with natural fillers derived from plant fibers (Mohanty *et al.,* 2002). The natural fibers like jute, flax, hemp, banana, sisal and

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coconut form a good source of natural fiber and alternative to synthetic fibers they also reduce dependency on non-renewable resource and lower pollution and greenhouse gas emission (Akindapo *et al.,* 2014). The natural fibers as fillers offer some advantages like low cost, high toughness, corrosion resistance, low density, good specific strength properties, reduced tool wear and biodegradability.

Matrices

The role of matrix in a fiber-reinforced composite is to bind the fiber together and transfer stress between the fibers, make them resistant against adverse environment and to protect the surface of the fibers from mechanical abrasion. The major matrices include Polymeric, Metallic, Ceramic and Carbon. Polymer matrix composites are most widely used material in industry. Polymer resins have been divided broadly into two categories: Thermosetting and Thermoplastics.

Thermosets are stiff and do not stretch they undergo irreversible solidification with processing temperature to about 200ºC. Commonly used thermosets as matrices for composites include resins like unsaturated polyester resins, Vinyl Ester, Phenolic Epoxy, Novolac and Polyamide (Hull and Clyne, 1996). Thermoplastics soften upon exposure to heat and return to their original condition at room temperature (Marsh and Bugusu, 2007). The thermoplastics whose processing temperature does not exceed 230°C are used as matrix as higher temperature causes degradation of fibers. The main disadvantages of using thermoplastic as matrix are high processing temperature, high viscosities, high coefficient of thermal expansion and they generally do not resist heat as like thermosets.

Epoxy resin

The word epoxy has come from epoxides; the epoxides are cyclic ethers (where an oxygen atom and two alkyl groups are in cycle). The common epoxide is “epichlorohydrin” it is the starting material of epoxy resin. The epoxy resin is synthesized by condensation polymerization reaction between epichlorohydrin and bisphenol (those which contain two hydroxyl phenol groups) there are different variants in bisphenol. Generally in condensation polymerization reaction water is eliminated but in this reaction hydrochloric acid is eliminated. The final product has epoxide group at the corners only which deliver them specific properties. The epoxy resin has numerous applications such as in paints/ coatings, adhesives, insulation in electronic goods and as matrix in composite materials. Epoxy comes in two parts system a laminating resin and a hardener. In a typical epoxy system, each molecule of hardener combines with four molecules of resin to form a three dimensional structure. Molecules also form hydrogen bond with each other and these lock adjacent sections of matrix and turn the matrix into more rigid structure. The epoxy resin can be classified based on their application such as civil application and laminating applications.

Natural fibers

The increasing pressure to reduce the utilization of plastic form various environmental board has motivated many academic and industrial persons to deviate towards, the use of natural fibers for the reinforcement of composites. Fibers can be obtained from primary plants which are grown for their fiber content such as Jute, Hemp, Kenaf and Sisal or secondary plant source where fibers are produced as a by-product like Pineapples, Baggase, Oil palm, Coir and coconut shell (Balaji *et al*., 2014). The natural fibers are renewable, cheap, recyclable and biodegradable. Their availability, renewability, low density and price as well as satisfactory mechanical properties make them an attractive ecological alternative to synthetic fibers used for the manufacturing of composites. The natural fiber composites are used in transportation (automobiles, railway coaches, aerospace), military applications, building and construction industries (ceiling panelling, partition boards), packaging, consumer products, *etc* (Chanap, 2012). Various natural fibers are used as reinforcing material in both thermoset and thermoplastics composites. However this paper reviews about coconut shell utilization as natural filler in composites.

Coconut shell

Coconut (Cocos nucifera) is a member of the palm family. Coconut shell is non-food part of coconut, which is hard lignocellulosic agro-waste. Coconut shell accounts to 15–20% of whole coconut (La Mantia, *et al.,* 2005). Coconut is mostly grown in tropical countries like India, Malaysia, Indonesia, Thailand and Srilanka. Coconut shell powder reflects to be a potential candidate for the improvement of new composites because of their high strength, modulus properties and cost effective (Vijayaram, 2013). It has good thermal stability compared to other agricultural waste. Different avenues of CS utilization are more or less known, but none of them have so far proved to be economically viable or commercially feasible (Madakson, 2012). Coconut is a fibrous polymer and they are classified based on whether the fibers are short or continuous. The Continuous fibers are more efficient at resisting loads than short ones, but it is more difficult to fabricate complex shapes from materials containing continuous fibers than from short fiber or particle reinforced material.

The methods used to produce fiber reinforced composites are similar as those of conventional molding methods used by industry. For thermoplastic methods include injection molding, extrusion, calendaring and Thermoforming. For thermosets methods include compression molding or matched die molding.

Chemical composition of coconut shell

The coconut shell fibers are basically rigid, crystalline cellulose microfibril-reinforced with amorphous lignin and/ or with hemicellulosic matrix. Most plant fibers, except for cotton, are composed of cellulose, hemicellulose, lignin, waxes and some water-soluble compounds, where cellulose,

hemicelluloses and lignin are the major constituents (Bledzki and Gassan, 1999). Hemicellulose is responsible for the biodegradation, micro-absorption and thermal degradation of the fiber as it shows least resistance, whereas lignin is thermally stable but prone to UV degradation.

The high lignin content makes the composites made with these filler more weather resistant and hence more suitable for application as construction materials. The shells also absorb less moisture due to its low cellulose content Table 1.

Table 1: Composition of CSP.

Attributes	% compostion
Moisture	7.073
Lignin	29.35
Cellulose	24.20
Hemicellulose	38.56
Ash	0.68

(Liyanage and Pieris, 2015).

Chemical treatment

The coconut shell as lignocellulosic filler exhibits various advantageous properties compared to synthetic filler. However, due to the presence of strong polarized hydroxyl groups on the surface of lignocellulosic fillers, the formation of a strong interfacial bonding with a non-polar polymer matrix becomes complex. The presence of pectin and waxy materials along with hydroxyl group tend to prevent the wetting of the filler surfaces. As a result, lignocellulosic filler composites suffer severe reduction in mechanical properties and delamination (mode of failure of composite where separation of layers occurs) (Taj *et al.,* 2007).

The interfacial adhesion among filler and matrix can be enhanced by surface modification of filler. Currently there are many methods to promote the interfacial adhesion between the lignocelluloses filler and polymer matrix, such as alkaline treatment, esterification, silane treatment, using compatibilizers and treatment with other chemical compound.

Alkalization/ mercerization

Strongly polarized hydroxyl groups make the cellulosic materials hydrophilic and chemically incompatible with the hydrophobic polymer matrices. Furthermore, the presence of wax, natural fats and cementing materials which may hide reactive sites and represents a hydrophobic blockage for fiber wetting and they must be removed. Alkali treatment, also called mercerization is the process of subjecting a natural fiber to the action of fairly concentrated aqueous solution of a strong base so as to produce great swelling with resultant changes in the fine structure, dimensions, morphology and mechanical properties. The treatment of the cellulosic material causes changes in physical structure and crystallinity of the fibers owing to its bleaching action (Das and Chakraborty, 2006). Bleaching removes the waxy and cementing substances like hemicellulose and lignin, resulting in the development of rough surface topography and breaking down of the composite fiber bundle into smaller and thinner fibers. The removal of these inert materials leads to new reactive sites that result in better interfacial adhesion

$$\text{Fibers-OH} + \text{NaOH} \rightarrow \text{Fibers-O-Na}^{+}\ \text{H}_2\text{O}$$

Many studies have been focused on alkali treatment of the natural fibers to enhance the interfacial adhesion between the fiber and matrix polymer (Cao *et al.,* 2006; Gomes *et al.,* 2004). Das and Chakraborty, (2008) noticed a steady increase in mechanical properties of bamboo strips with increasing concentration of NaOH.

Oxidizing agents

The use of oxidizing agents, such as hydrogen per oxide (H_2O_2) is another effective way to remove the cementing materials from the natural fiber that hinders its adhesion with polymer matrices. The reaction is given below

$$H_2O_2 = H^+ + HO_2^-$$
$$HO_2^- = OH^- + [O]$$
$$[O] + I = I$$

(Where; I = Impurities)

Alkali treatment of the natural fiber may lead to formation of alkali resistant linkage between lignin and hemicellulose. This linkage impedes the removal of lignin. The use of H_2O_2 can break this linkage and delignify lignocellulosic fiber to enhance its interfacial adhesion with the polymer matrix. Modibbo *et al.,* (2009) used H_2O_2 treatment as precursor to NaOH treatment and found that the treatment has reinforced the fiber and consequently improved its crystallinity and strength. In an alkaline condition, the hydroxyl ions (OH^-) produced by the perhydroxyl ion HO_2 hydrolyses the hydrogen ions (H^+), thereby promoting the liberation of more perhydroxyl ions.

Coupling agents

The use of coupling agents is another effective way to mitigate inherent dissimilarities and to enhance interfacial adhesion between the fiber and the polymer matrix. The coupling agents have bi-functional groups that are capable of reacting respectively with the fiber and the matrix. Organofunctionalsilanes belong to the most widely used group of coupling agents having a generic chemical structure R(4-n)-M-(R^1 X)n (n = 1,2) where R^1 is alkoxy (hydrolysable) group, M is tetravalent base metal (Si or Zr), X represents an organofunctionality and R^1 is an alkyl bridge connecting the base metal and the organofunctionality material (Huda *et al.,* 2007; Xie *et al.,* 2010). Alkoxy groups are hydrolysable and they undergo hydrolysis in the presence of moisture which causes the formation of silanols. The silonal then react with the hydroxyl groups of the fiber, forming stable covalent bonds to the cell wall that are chemisorbed onto the fiber surface (Agrawal *et al.,* 2000; Li *et al.,* 2007). This chemisorption usually improves the degree of cross linking in the interface region and improves chemical affinity to hydrophobic polymers. Furthermore, the presence of alkyl group (R^1) in presence of silanes, influence the wettability

of the fiber and reduces its water uptake because of the crosslinking due to covalent bonding between the matrix and the fiber.

Researchers on the basis of contact angle measurements of the chemically treated samples, have concluded that the modification has resulted in a change from highly hydrophilic polar surfaces to hydrophobic-totally non polar surface. There have been some studies reporting the enhanced mechanical properties and reduced water absorption of the natural fiber reinforced biocomposites, on incorporation of silane treated fibers, resulting from the improved adhesion between the fiber and matrix (Lee *et al.,* 2009; Herrera-Franco and Valadez-Gonzalez, 2005).

Acetylation

Acetylation of the natural fiber is another effective way to improve its hydrophobic characteristics and thus enhance their interfacial adhesion with the matrix and reduce its moisture uptake. It involves the use of acetic or propionic acid at elevated temperatures with or without an acid catalyst. Acetylation of the cellulosic material results in the substitution of the hydroxyl groups of the cell wall with acetyl groups. As a result, hydrophobic tendency of the natural fiber improves and hence enhancing the compatibility with polymer matrix and reducing the water absorption tendency.

Compatibilizers

Due to their compatibility with both, the filler and the matrix, they are termed as compatibilizer. The compatibilizer is usually a graft copolymer of the polymeric matrix and an anhydride such as maleic anhydride (MA) (Bendahou *et al.,* 2008). These regents are compatible with the polymer matrix and the hydroxyl groups of the lignocellulosic material, forming covalent bonds. It is widely used method for the production of natural fiber reinforced polyolefin composites like Polyethylene and Polypropylene (Feng *et al.,* 2011; Keener *et al.,* 2004).

Coconut shell powder based epoxy composites

The incorporation of filler such as coconut shell powder into thermosetting material enhances mechanical properties and also reduces the production costs of the molded products. The natural fiber based epoxy composites are been extensively promoted by manufactures in making automobiles components, building, marine, packaging, sporting electrical and even in manufacturing of aircraft components. Nowadays fiber reinforced epoxy composites are widely used in orthopedics and artificial limbs. In dental applications, composite resins are fully replacing the conventional restorative materials such as amalgam, gold, alumina and zirconia. In coming day's fiber reinforced composites will be in high demand (Saba *et al.,* 2016). Some of the studies conducted on CSP based epoxy composites are discussed below Fig 1.

Vignesh *et al.,* 2015 studied the effect of addition of CSP as filler in epoxy composites, the CSP was treated in sodium hydroxide (NaOH) and potassium hydroxide (KOH) with 2%, 5% and 8% concentration at constant soaking period of 24 hr. The weight fraction of composites was maintained at 30% particle and 70% remaining resin, hand lay-up technique was used for production of composites. He observed that the mechanical properties of the composites such as flexural strength, impact strength and hardness improved. Ozsoy *et al.,* (2014) used chopped bamboo and chopped coconut shell as reinforcement material at three different weight fractions (6%, 8% and 10%). The matrix used in composite was epoxy resin (MGS L285) mixed with hardener (HGS L285) in mass ratio of 100/40, which was mixed with reinforcement material to manufacture composites. He observed that the tensile and flexural strengths of composites were affected by the amount of reinforcement in the composites. The bamboo reinforced epoxy composites were stronger than coconut shell reinforced composites. Hardness of the composites increased with increasing of reinforcement.

Muthukumar and Lingadurai (2014), developed epoxy matrix composite using coconut shell powder (CSP) and groundnut shell powder (GSP) in different volume as reinforcing material. The composites were moulded in a mould with the dimension of 300 mm × 300mm × 3mm and cured at temperature of 80°C - 120°C. During investigation he observed that the mechanical behavior *viz*. Tensile strength, flexural strength, Impact strength of CSP and GSP epoxy composite Material was greatly influenced by the CSP and GSP filled volume fraction. The maximum tensile strength is obtained for the composite prepared with 40% CSP and GSP volume fraction. However with further increase of filler volume the tensile strength goes on decreasing. The maximum flexural strength was obtained

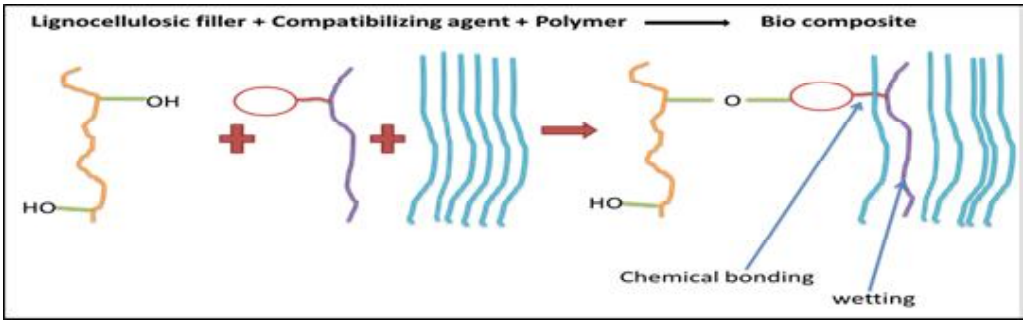

Fig 1: The function of compatibilizing agent in the lignocellulosic fiber reinforced polyolefin composite.

for the composite prepared with 50% CSP and GSP filled while the flexural strength decreases on increasing filler volume from 50% to 60%. Singh *et al.,* (2014), used coconut shell powder in different particle size for reinforcing in different volume in epoxy matrix. Epoxy resin Moditite EL 301 of medium viscosity and Hardener MH-933 were used to harden matrix were used. He observed that the maximum tensile strength is obtained for the composite prepared with 20% CSP volume fraction. The maximum flexural strength is obtained for the composite prepared with 30% CSP filled composite whereas the minimum flexural strength is obtained for the composite prepared with 40% CSP filled composite. Kumar *et al.,* (2014), developed CSP reinforced epoxy matrix composite. Four sets of composites with 20% volume fraction of coconut shell powder with different combination of stirring parameters were prepared and ultimate tensile strength (UTS) was evaluated. The epoxy and hardener were mixed in the proportion 2:1 by volume and using a mechanical stirrer the mixture is stirred vigorously. The effect of stirring on the UTS of composites was studied. He observed that, addition of hardener at the beginning of stirring causes improper mixing and results in poor UTS. The mixing at higher temperature of epoxy, coconut shell particles and hardener also showed highest value of UTS due to homogeneous mixing at higher temperature. And as hardener was added at high temperature, there is no scope for the particles to settle down and deteriorate the tensile properties. Bhaskar and Singh, (2013), developed Coconut particle reinforced composites by reinforcing shell particles of size between 200-800 ìm by wt% of 20, 25, 30 and 35 into epoxy matrix. Experimental results showed that density, ultimate strength, modulus of elasticity and % elongation decreases with wt% of shell particle. Tensile strength of 25 MPa and modulus of elasticity of 654 MPa were retained even after of 35% reinforcement. Properties were comparable for application only with compromising slightly with matrix property.

CONCLUSION

The demand made by the industry for materials that are both light and strong has been the main driving force for the development of composites. There is an immense opportunity to utilize the agricultural waste mainly fibers for the reinforcement in polymer matrices to obtain low cost composites. The use of Natural fiber in polymer composites, are alternate to inorganic fillers. It is of great interest in the view of the reduction in the use of petroleum-based, nonrenewable resources and in general it is the more intelligent utilization of environmental and financial resources. The coconut shell powder has emerged has one of the good natural filler for biodegradable environmental friendly composites. It has Good thermal stability compared to other agricultural waste, can be easily crushed into particles and generates rural jobs and improves standard of living in rural areas. The natural fiber reinforced composite find application in various industries, due to their several beneficial properties however they to have their own weakness which are overcome by chemical modification of fillers or use of adhesion promoters and additives. There has been a lot of research work done on coconut shell based composites, but there is lot of scope to improve the properties and overcome the difficulties. The commercial situation of natural filler based composites in still in nascent stage, so much work can be done to improve their properties and to find place for these materials in market. As soon as the market for these composites increases, reduction of costs and improvement of the quality will be achieved.

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