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Natural Filler based Composites: A Review

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Abstract : Due to concerned global environmental issues and better mechanical properties, fibre reinforced composites are gaining popularity in last few decades. Natural fibers have been successful in imparting to composite material certain benefits like low density, reduced machine wear compared with synthetic reinforcement materials and a lowering of associated health hazards; hence the considerable amount of research is directed towards using lignocellulosic fibers from a wide range of sources as a reinforcement component. This paper deals with the review of various researches carried out to study the effect of natural fillers on the mechanical properties of the polymer composites.

Keywords: Natural Fillers, Polymer Composites, Bio-composites, Mechanical Properties .

INTRODUCTION

Natural fibres by virtues of its advantages like low cost, low density, availability in abundance, environmental friendly, non-toxicity, high flexibility, renewability, biodegradability, relative non-abrasiveness, high specific strength and modulus and ease of processing, proves to be better alternative than synthetic fibres[1-8]. Natural fibres comprises of cellulose, hemicellulose and lignin [9, 10]. Natural fibre-reinforced composites are successfully used for various commercial applications such as deck surfaces, door components, windows, packaging and automotive industries and furniture [11, 12].

In general, composite consists of two distinct components, fillers and matrix, which once joined together produces a totally different material having properties different from those of the constituent materials. The reinforcement or the filler phase consists of stiff, strong material, fibrous nature, embedded into a continuous matrix phase. The advantage of such a composite is that high strength and stiffness of the fibres may be exploited [13]. Natural and Regenerated cellulosic fibres are one of the most important aspect of bio-composite reinforcement is discussed in this paper. Following researchers have investigated different aspects of filler matrix interactions and the effect of fillers on the different properties of the composites.

Ofora et.al. [14] have studied the effect of varying quantities of animal based fibres like feather, hide and hoof on the physico-mechanical and thermal properties. The composites were prepared using the injection molding technique in which the premixed amount of polystyrene pellets and calculated amount of filler particles of size 200 μm were fed into the machine and test samples were prepared. These researchers have concluded that the fillers, hide and feather exhibit increased tensile strength while hoof filler have shown a decrease in the tensile strength as compared to the unfilled specimen. This was attributed to the degree of adherence of the filler particles to the polymer matrix. A tremendous increase in compressive strength is observed which was because of better polymer-filler interaction and adhesion resulting in the stiffening of polystyrene chain and therefore a resistance to compression under applied strain. In case of flexural strength, hide and hoof fillers have better interaction with polystyrene matrix, thereby resulting in better flexural strength. Also as the amount of filler is increased, both melting and crystallization temperatures are increased as compared to unfilled polymer.

Nwanonenyi et.al [15] have investigated the effect of particle size, filler content and compatibilization on the properties of the low density polyethylene composites filled with periwinkle shell powder on some of the mechanical properties. Different particle sizes (75,125 and 150 μm) are used at different fiber loading (0-30%) with and without the use of compatibilizing agent (Maleic Anhydride) were employed for this study. The results indicated an increase in tensile strength, flexural strength, impact strength and specific gravity as the filler loading and compatibilizer content is increased for all sizes of particles. This enhancement in the property is attributed to the better interaction between the filler particle and the matrix.

Atluri et al. [16] have studied the mechanical properties of golden cane fibre and polyester composites. Composites were prepared using hand lay-up method and cured in the mold to prepare the specimen for testing. The fiber volume fractions were varied to study the effect on tensile strength, flexural strength and impact strength of the prepared composites. It has been concluded that the golden cane fibres improve the tensile strength, stiffness, flexural strength, modulus and toughness of the resin alone and reduces its density with increase in volume fraction of fibres in the composite.

Sakthivei and Ramesh [17] have studied the effect of fibers like coir, banana and sisal on the properties of the synthesized epoxy resin composite. Hand layup method is used to prepare the samples in the designed mold. The fibers used in the composite preparation is washed and chemically cleaned for the composite preparation. The results concluded that among the tested composites the banana fibres composites have shown the best enhancement in properties as compared to sisal and coir reinforced composites.

Jagadeesh et al. [18] have investigated the effect of filler size and loading of corn husk flour filled polypropylene composites. The composites are synthesized by the process of extrusion and injection moulding. It has been observed that the flexural and impact strength decreased with increasing filler content but enhanced with decreasing particle size. This was attributed to the fact that lower filler size offers larger specific surface area than those of larger mesh size at same weight fraction. Verma D. K. et al. [19] have studied the effect of natural fillers (rice husk, wheat straw and wood flour) on impact property of GPSS composites. The impact strength in general was increased with the addition of fillers. It was concluded that the dispersion was not optimized and further improvement of the processing conditions or use of a compatilizer is needed to find the best possible dispersion of lignocellulosic natural fillers with GPPS matrix.

Hardinnawirda and Aisha [20] have studied the effect of filler loading on the mechanical properties of rice husk filled unsaturated polyester resin composite. The samples were prepared by molding method. It was experimentally justified that as the filler loading is increased the tensile strength is decreased but at 25% filler content there is slight increase in tensile strength. This has been attributed to the fact that the weak bonding of the hydrophilic filler and the hydrophobic matrix restricts the stress propagation and tensile strength is decreased. Also poor dispersion causes filler agglomeration. At high filler content, there was lack of resin that can wet the filler which results in lesser stress to be transferred thereby increasing tensile strength at 25 % filler loading.

Thakur et al [21] have studied the effect of filler loading on green composites using Grewia Optiva fibres as filler fibers and Phenol formaldehyde as matrix on the mechanical and chemical aging properties of the synthesized composites. These composites are prepared using molding method. Tensile, Compressive and flexural strength increases with increase in filler loading. In case of wear resistance, the reinforced polymer shows better wear resistance as compared to virgin polymer. It was also observed that as the filler loading is increased the swelling behaviour in water has also increased. For chemical resistance the higher filler loading makes the composite more prone to damage.

Wollerdorfer and Bader [22] have studied the influence of plant fibres on the mechanical properties of polymers using thermoplasts like polyester, polysaccharides and blends of thermoplastic starch. The composites are prepared using extrusion and injection molding process. On the basis of experimental results it was concluded that a considerable strength improvement is not observed in case of polyesters while in case of polysaccharides, the chemical similarity of matrix and filler has resulted in the increase in tensile strength. A considerable increase in tensile strength is observed for the composites reinforced with wheat starch and a blend of cellulose diacetate and starch blends.

Khan, Mubbasher Ali and Siddiqui M. Arif, [23] have studied the effect of filler loading on the talc reinforced polypropylene composite. Talc added PP composites showed an increase of modulus of elasticity upto 20 percent addition of talcum powder as compared to the pure PP and decreased at higher concentration. Small increment in modulus of elasticity was also found with increase in injection pressure. Sapuan and Bachtiar [24] have studied the tensile strength of sugar palm fibre reinforced high impact polystyrene composite. Different fibre loadings were utilized for investigating the effect of filler on the properties. The investigations concluded that increase in filler loading improved the tensile strength and tensile modulus of the composite.

Onuegbu and Igwe [25] have studied the mechanical properties of snail shell powder filled polypropylene composites. The particle size and filler loading were varied to study the properties of the composites. These composites were prepared using injection moulding machine and later extruded as sheets. The results have shown that the filler increases the tensile strength, flexural strength and impact strength with increase in the filler content and decease in the filler size. However the elongation at break was observed to decrease with increase in filler content and particle size. Also the water absorption, hardness and specific gravity of the composite increase with increase in filler content and decrease in filler size. Cerqueira et al. [26] have studied the mechanical properties of the chemically modified sugarcane bagasse fibre and polypropylene composite. The filler particles were pre-treated with sulfuric acid followed by lignification with sodium hydroxide solution. These filler particle are mixed with PP in a thermokinetic mixture and the specimen were prepared by injection moulding process. An effective improvement in tensile, flexural and impact properties were observed during the investigations.

Souza et al [27] have studied the effects of pre-treatment of textile fibre on the mechanical properties of textile fibre filled HDPE composites. The composite was prepared using thermo-kinetic mixer and the specimens were prepared for tensile test and flexural test using injection moulding. The results have concluded that filled composites have better mechanical properties as compared to the virgin HDPE. Ahmed Shahbaz et al. [28] have prepared polystyrene composite using in-situ polymerization technique having four different inorganic fillers i.e. Fillers calcium carbonate, graphite, mica and talcum powder with three different filler loading. The melt flow index was performed to study the effects of Fillers and their concentrations on the flow properties of composites. The morphological analysis by SEM and EDX were also performed. The particle size was found to be varying from 0.2 μm to 0.5 μm.

Zizumbo et al [29] have studied the effect of modified sugarcane bagasse fibre on the mechanical properties of the polystyrene composite. In this work, the bagasse fibres were modified using dichloromethylvinylsilane and grafted with polystyrene were studied to study the effect on the properties of fibers and composites. FTIR was used to characterize the fibers while TGA is used to study the thermal properties while SEM is used to study the surface of fractured composites. The results concluded that silanized and polystyrene grafted fibres have resulted in better properties of the composites as compared to the untreated fibres.

Zaini M.J. et al. [30] have investigated the effect of filler content and size on the mechanical properties of the wood-based filler, oil palm wood flour in polypropylene. Four different sized fillers were compounded using a twin screw compounder. As filler content increased, the mechanical properties decreased. The larger sized filled composites showed higher modulus, tensile and impact strengths particularly at high filler loadings. Ibrahim M.S. et al. [31] have investigated the effect of different contents of oil palm ash filler on the mechanical and thermal properties of unsaturated polyester composite. It was concluded that the mechanical properties like tensile and flexural strength were improved with increasing filler content. Also the thermal stability increased as the filler content increased. The SEM results were extensions of these results.

Rozman H.D. et al. [32] have investigated the effect of filler size and coupling agents on the composites made from rubberwood fillers in the form of fibers (RWF) and powders (RWP) and high density polyethylene (HDPE). The RWP-HDPE composites showed higher tensile strength than those of the fibers. 3-(trimethoxysilyl) propyl methacrylate (TPM) and 3-aminopropyltriethoxysilane (APE) were employed as coupling agent to study the mechanical properties. TPM significantly improved the modulus of elasticity (MOE) and the impact strength of the RWF-filled composites, however it resulted in reduction of tensile modulus and increase in elongation at break for both RWF and RWP-filled composites. On the other hand, APE produced RWP-filled composites with a higher tensile strength and modulus.

Ahmed Shahbaz et al. [33] have synthesized polystyrene composites filled with four fillers calcium carbonate, graphite, mica and talcum powder with three level of concentration 1%, 2%, 3% by weight. The hardness and wear tests were performed to study the effects of fillers and their concentrations on the composites. The maximum improvements 102.08% in synthesized PS-$CaCO_3$ composite and 62.83% in case of synthesized PS-talcum powder composite for hardness and wear resistance were observed. The addition of fillers are found to be significant while concentrations of fillers insignificant at 5% level of significance.

Premlal Hattotuwa G.B. et al. [34] has studied the effect of filler type and filler loading on mechanical properties of polypropylene composites. Ground talc and rice husk were used as filler. It was concluded that as the filler loading increased Young’s modulus and flexural modulus increased, whereas yield strength and elongation at break decreased for both types of composites. Rice husk composites exhibited lower yield strength, Young’s modulus, flexural modulus and higher elongation at break than talc composites.

Hatta N. and Akmar N. [35] have studied the effect of addition of different ratios of coconut and jute fibers on mechanical properties of polystyrene/polypropylene composite. The addition of 10% wt of fiber has increased tensile properties of composites. The tensile strength of 100% jute fiber reinforcement ratio was higher than 100% coconut fiber reinforcement, while on the other hand the value of young’s modulus in which composites reinforced with 100% coconut fiber showed the highest value and decreased with increase in jute fiber. Also the impact strength of the composites with 10% fiber is smaller. Ahmad I. et al. [36] have investigated the effects of alkali treatment, filler content, and filler size on the tensile, flexural, hardness and water absorption for the sawdust filler composites with unsaturated polyester resin which was prepared by recycling of polyethylene terephthalate (PET) waste bottles. The investigations concluded that as the filler contents increased the tensile and flexural moduli also increased but the tensile and flexural strength decreased. Also smaller size saw dust produces higher strength and modulus. It was also concluded that alkali treatment causes a better adhesion between sawdust and polymer matrix.

Ahmed Shahbaz et al. [37] have studied the tensile properties of synthesized polystyrene composites using calcium carbonate, graphite, mica and talcum powder as fillers with different loading. Maximum increase of 72.50% in synthesized polystyrene-talcum powder composite and minimum increase of 10.35% in case of synthesized polystyrene-mica composite were observed. The order of maximum improvement in tensile strength of synthesized polystyrene is found to be mica powder < graphite powder < $CaCO_3$ powder < talcum powder.

Maheshwari C. Uma et al. [38] have studied the tensile and thermal properties of Polycarbonate coated tamarind fruit fibers. The tensile properties of alkali treated and polycarbonate coated tamarind fiber have better tensile properties owing to the better interfacial bonding between the fiber and the matrix. It was also concluded that the thermal stability of polycarbonate coated fiber was higher than that of the uncoated fiber but lower than polycarbonate. The moisture absorption of polycarbonate-coated fiber was lower than that of uncoated fiber. Mehta N.M. and Parsania P.H. [39] have fabricated hybrid composite based on Bisphenol-C-formaldehyde resin and jute mat with rice, wheat, sugarcane and jamun husk. The resin content in composites was 50% of fibers. Tensile strength, flexural strength, electric strength and volume resistivity of hybrid composites have been investigated and compared with those of jute-bisphenol-C-formaldehyde composites. It is observed that tensile strength of composites decreased by 53-72%. Flexural Strength has increased by 53-153% except jute-rice husk composite for which it is decreased by 26%. A little change in dielectric breakdown strength (1.89-2.11 kV/mm) is found but volume resistivity of jute-wheat husk and jute-jamun husk composites has improved by 437-197% and decreased slightly (2.3-25.2%) for the remaining two composites

Khan. Md. Aiyaz Ali., et al. [40] have synthesized the sugarcane bagasse (SCB) reinforced polystyrene (PS) composite and studied the effect of different fiber sizes on the hardness of the prepared composite. It was observed that as the size of the filler decreases hardness increases. This may be due to the fact that the dispersion of the SCB particles in the PS matrix is better than the other particle sizes used. Also better dispersion of filler particles is attributed to the better hardness of the composite. SEM analysis shows that as size decreases, dispersion of filler improves in polymer matrix.
Binhussain Mohammed A. and El-Tonsy Maher M. [41] have produced an environmental friendly artificial wood for structural applications, by recycling mixed plastic waste and date palm leaves waste. The density, water absorption, hardness, modulus of elasticity(tensile and flexural), impact strength and linear burning rate of different wood plastic composites namely polycarbonate, polystyrene and polyvinyl chloride were recorded and compared with the natural hard, soft wood and medium density fiberboard woods. The developed composite exhibited less water absorption, linear burning and hardness, higher density than that of natural and medium density fiberboard and is feasible for outdoor usage.

Digabel F. Le et al. [42] have produced a composite from lignocellulosic fractions from wheat straw were used as natural fillers in the matrix of polyolefin and a biodegradable polyester [poly(butylenes adipate-co-terepthalate)]. Investigations of tensile and impact properties revealed a reinforcing effect for both types of composites. The polyolefin composite were found to be more brittle as compared to polyester based composites. Also addition of compatibilizing agents did not improve the properties of polyolefin composites.

Raju G.U. et al. [43] have investigated the properties of groundnut shell particles reinforced vinyl ester resin composites. Different weight percentages of particles in polymer matrix were investigated and it was observed that maximum rupture modulus at 20% reinforcement and maximum elastic modulus is obtained at 60% reinforcement while maximum tensile strength and maximum Young's modulus is observed at 40% reinforcement. Impact strength shows a steady increase in impact strength upto 50% of filler addition. Moisture content is found to be 1.92-4.96% and water absorption was only 1.51-8.82%. Nassar Amal and Nassar Eman [44] have studied the effect of nano SiC particles on the mechanical particles for the epoxy resin composite. It was experimentally concluded that tensile strength decrease with increase in weight percentage of reinforcement. The wear results show that nano particles enhanced the wear resistance of composite.

Yildiz Ayse Betul and Cetinkaya Kerim [45] have produced a polymer composite specimen using glass fiber, polyester reinforced banana fibres and PET bottles and performed tensile and impact tests to study the properties of the composite. In the light of above experiments, it is concluded that agricultural wastes offer a potential alternative raw material for the forest industry. Moresco Mauro et al. [46] have prepared agro filled polymer composite containing six different agro fillers were prepared and their properties were examined in terms of the density of the fillers and the composites. The composites showed decreased tensile strength and elastic modulus with filler loads and specific mechanical properties showed similar trends.

Mosawi Ali I. et al. [47] have evaluated the mechanical properties of araldite matrix composites reinforced with palms- Kevlar fiber with different reinforcement percentages. The results show the improvement in the impact strength, tensile strength, flexural strength and hardness with increasing percentage of reinforcement. Khan. Md. Aiyaz Ali. et al. [48] have studied the effect of different fiber size on mechanical properties of the sugarcane bagasse fibers (SCB) reinforced polystyrene composites. Addition of filler particles improves the mechanical properties and as the size of the particle decreases, tensile strength increases. With increase in filler size, flexural strength increases upto a level and then decreases. Composite having filler size 105μm to 150μm depicts a significant increase in flexural strength. The effect of particle size on wear rate is significant. The lower the particle size, the lesser would be the wear rate at all the loads studied.

Rosa Simone Maria Leal et al. [49] have prepared the polymer composite using rice husk flour (RHF) as filler in the matrix of polypropylene using melt extrusion. Maleic anhydride modified polypropylene (MAPP) was added as a coupling agent. The tensile strength decreased as the filler loading is increased. The presence of MAPP improved the tensile strength showing a strong independence on MAPP/RHF ratio. The density of the composites slightly increased with filler and coupling agent in comparison to pure polypropylene. Also the presence of MAPP diminished more than 20% intake in highly loaded composites.

Danesh Mohammad Amin et al. [50] have investigated the morphological and thermal properties of waste newsprint/recycled PP/nanoclay composite. The tensile properties of the blended composites with the addition of nano clay were further examined using SEM and XRD. Thermal degradation behavior of the composite showed that the degradation temperature shifted to higher values after addition of nanoclay. The experimental results concluded that these waste materials could be used as appropriate alternative raw materials for making low cost wood-plastic composites,
Gupta Nikhil et al. [51] have investigated the effect of addition of fly ash as filler on the compressive and impact

properties of glass fiber reinforced epoxy. It was concluded that compressive strength of the material is found to decrease whereas steep increase in impact strength is observed by introduction of very small quantities of fillers. Effect of specimen ratio on the compressive strength was also studied and significant effect was observed.

Raja R. Satheesh et al. [52] have studied the effect of fly ash filler size on mechanical properties of polymer matrix composites using epoxy polymer as matrix. It was experimentally concluded that the size reduction of fly ash particle enhanced the strength of composite. Yao Fei et al. [53] have investigated the mechanical properties and crystallization behavior of composite of virgin and recycled high density polyethylene and five types of natural fiber fillers i.e. four types of rice husks (rice husk, rice straw leaf, rice straw stem and whole rice straw) and wood fibers. It was experimentally concluded that for both virgin and recycled, rice straw systems had comparable mechanical properties with those of wood composites. Increase in filler loading led to increased moduli and decreased tensile and impact strength. It was also observed that with rice husk had the smallest storage moduli, but their impact strength was comparable or better than that of other straw fibers. Very little difference was found to exist in mechanical properties for different rice fibers. The particular recycled HDPE resin and its composites have better moduli and strength properties compared to the virgin HDPE. The study concluded that rice straw fibers can work well with both virgin and recycled HDPE.

Salaman Ali J. [54] has studied the tensile and impact properties of polystyrene matrix composite reinforced with palm natural fibers and carbon fibers. The maximum tensile strength (358 MPa) was found at the weight fraction of 60 % of fibers as compared to (59 MPa) for virgin polystyrene. The maximum impact strength is 175 kJ/m2 at weight fraction of 60 % of fibers as compared to (19 kJ/m2) virgin polystyrene.
Aridi et al [55] have studied the effect of filler loading and coupling agent on the physical, mechanical and morphological properties of the rice husk filled polypropylene composites. The struktol coupling agent was used to help the interaction between the filler and the matrix. The results concluded that 50% filler loaded composites had optimum flexural strength, flexural modulus and tensile strength whereas the 35% was the best filler loading for Young's modulus, flexural strength, flexural modulus and impact strength.

CONCLUSIONS

This paper deals with the review of the literature pertaining to the effect of different fillers on the mechanical properties of the thermoplastic composites. The researches have collectively focused on how the addition of fillers helps in improvement of the mechanical properties and on the other hand results in the more economical alternative, the unfilled polymers. These researches have also given the direction for future research with respect to the surface modifications of the fillers, different matrices that can be used for composites preparations, fabrication methods that are used for preparation of composites and specimens to be tested.

Further research is required for the composites to overcome the environmental degradation, moisture absorption and the detrimental effects to the aesthetics and performance of the composites when exposed to the temperature, humidity or UV radiation. These performance obstacles have resulted in the handicap of these composites to be used widely for the industrial, engineering and commercial applications. A great amount of work is being done and more is required to be done in this area.

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