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Abstract: In modern years, both industrial and academic world are focussing their attention toward the development of sustainable composites, reinforced with natural fibres. Recent progress in the field of intrinsically conductive polymers (ICPs) as well as conductive polymer composites (CPCs) filled with natural fibres is reviewed. The possibilities of utilizing natural fibres as fillers for ICPs as well as CPCs to form natural fibers-conducting polymer composite materials have wide potentials in the modern industries. The unique characteristics such as electrical conductivity, mechanical strength, biodegradability and recyclability enabled them to be implemented in many novel and exciting applications including antennas, chemical sensors, tissue engineering, neural probes, biosensors, drug delivery, bio-actuators, fuel cells etc. The effects of fibre contents, fibre size, chemical treatment, temperature and moisture content on the dielectric properties of the conductive composites were reviewed.

Keywords : Natural fibres, conductivity, chemical compositions, fillers, reinforcement.

1. Introduction

Due to the growing social, economic and ecological awareness along with government emphasis on the environmental impact and sustainability, the proper utilization of natural resources and wastes are strongly encouraged [1-4]. Consequently, the natural fiber composites (NFCs) have become valuable alternatives for various industrial applications. In NFCs, natural fibres are used as fillers or reinforcing materials for polymer matrices [5-8]. The proper utilization of natural fibers not only resolves the waste disposal problems but also reduces environmental pollution [9-11]. NFCs are attractive from environmental point of view which enabled them to be used as an alternative to the traditional glass/carbon polymer composites [12-13]. They are used in various applications including packaging, furniture, automotive industries, disposable accessories, building, and insulation materials [14-15]. In addition, these NFCs show several advantages and superior characteristics over traditional composites due to the low cost and densities along with acceptable specific strengths and moduli [16-19] which offer the opportunity to produce light weight products. Furthermore, NFCs are also used in producing recyclable and bio-degradable products [20-23]. On top of that natural fibers have several advantages over glasses such as: availability, reduced tool wear in machining, CO2 sequestration enhanced energy recovery, and reduced dermal and respiratory irritation [24-27]. Despite of that, natural fibers suffer from certain considerable drawbacks like poor water resistance, low durability, and poor bonding with the matrix. This weak interfacial bonding leads to undesirable characteristics of the composites and thus affects their industrial usage [28-30]. Therefore, different solutions have been offered to improve their compatibility and bonding such as usage of coupling agents and surface treatments via mechanical, chemical, and/or physical modifications [30-35]. Natural fibers can be classified based on their origins such as bastfibers, leaf fibers, fruit,
and seed fibers. Wide range of natural fibers has been used to reinforce different polymer matrices. Such fibers include wood, bamboo, cotton, coir, rice straw, wheat straw, rice husk, flax, hemp, bagasse, pineapple leaf, oil palm, date palm, curaua, ramie, jowar, kenaf, doum fruit, rapeseed waste, sisal, jute etc. [36-40]. Fig. 1 shows natural fibre segments.

**Fig. 1 Natural Fibre Segments**

### 2. Polymer conductivity

In recent years, their potential applications in functional papers as well as packaging industries have drawn special attention. Several studies confirmed dire need of using such conductive polymers in electrical applications. Coated paper with conducting properties can be used to make anti-static and electro-magnetic shielding papers, anti-bacterial papers, novel wall coverings and electrical resistive heating papers [40-42]. Author [42] prepared conducting paper utilizing natural fibers and conductive polymers, where unbleached bagasse and/or rice straw fibers were infused into polyaniline (PANi). Differential scanning calorimeter (DSC), Fourier transform infrared (FTIR), spectroscopy thermal gravimetric analysis (TGA) etc. were used to characterize such produced composites, whereas scanning electron microscope (SEM) was used to investigate themorphology. Results showed that increased conductivity was obtained with the increase of PANi in the composite. However, the breaking length, tear factor and burst factor decreased with the increase of PANi, and such effects were more obvious in bagasse-based composites. In addition, the cure characteristics, thermal and microwave properties, DC conductivity as well as mechanical properties of both natural rubber/polypyrrole and natural rubber/polypyrrole/polypyrrole- coated short nylon fiber composite were studied [43]. It was noticed that the DC conductivity of the natural rubber/polypyrrole composite was enhanced only at very high polypyrrole loading and the maximum conductivity was achieved at 100 phr loading. Results also showed that the composite’s thermal stability was increased withloading of polypyrrole and polypyrrole coated fiber, whereas dielectric constant as high as 55.5 was obtained for 100 phrpolypyrrole loaded sample at 3.98 GHz frequency. The conducting composites showed substantial improvement in dielectric heating coefficient as well as skin depth and absorption coefficient. Moreover, conductive papers of graphite/carbon/cellulose fiber composites with low production cost, good mechanical properties and tunable electrical conductivity were produced [44]. It was observed that ultra violet absorbance was increased with increasing the Carboxymethyl cellulose. Furthermore, efficient conductive nano-filler pathways were made through agglomerates or dispersed nano-filler [45]. It was also concluded that resistivity dramatically differed with the dispersion of carbon nano-fiber in polycarbonate that were controlled by sonication conditions. Also, conductive polymer with silk fiber bundle was successfully utilized in making string-shaped electrodes [46], where electro- conductive polyelectrolyte, poly(3,4-ethylenedioxythiophene) -poly (styrenesulfonate) (PEDOT-PSS) and silk thread were combined in electrochemical manner to develop the electrodes. The polymer composite was shown to have a conductivity of 0.00117 S/cm. The addition of glycerol to the PEDOT-PSS silk thread was able to improve the conductivity to 0.102 S/cm.
3. Dielectric properties of fiber composites with fillers

It was also studied that composites with hybrid (CFF-E-glass/Epoxy) fibers usually had a low value of dielectric constant. On the other hand, highly conductive polymers with silk fibroin composite fibers were fabricated via in-situ polymerization by Xia and Lu [47]. It was reported that polypyrrole/silk fibroin, polyaniline/silk fibroin, and poly3,4-ethylene-dioxythiophene/silk fibroin composite fibers exhibited varied conductivity in the range of 3.8–4.2 respectively. It was also shown that these composites demonstrated better electrical and thermal characteristics and may have potential applications as novel functional materials in textile and biological areas. The electrical resistivity of composites based on polypropylene/coconut fibers composites was also studied [48]. They aimed to investigate the electrical properties of low cost and eco-friendly composites to improve their implementation in the industrial applications. On the other hand, the electrical conductivity of composites based on epoxy resin with polyaniline-DBSA fillers was studied and analyzed [49]. They have utilized all of conductive filler PANI-DBSA in form of powder and paste in matrix polymer bisphenol, hydride hardener and epoxy resin as well as accelerator to form the composite.

4. Effect of fiber content with various chemical compositions

Author [50] compared the electric characteristics of natural fiber reinforced/LDPE composites with glass/LDPE and carbon black/LDPE composites. It was reported that the small change of dielectric constant of glass/LDPE composite occurred with increasing frequency and fiber content compared to coir and sisal fiber/LDPE composites. It was also reported to be due to low interfacial polarization. Authors also reported that electrical conductivity of hydrophobic LDPE can be improved by mixing it with hydrophilic lignocellulosic fibers and conductive carbon black. Moreover, author [51] reported a change in the dependence of dielectric properties of short jute fiber reinforced polypropylene composites as fiber loading changes at critical fiber content.

Different chemical treatments like potassium permanganate, toluene di-isocya-nate (TDI), maleic anhydride modified polypropylene (MAPP), and stearic acid (ST) were applied to jute yarn/polypropylene composites [52] to enhance the interfacial adhesion between the matrix and the filler. It was noted that the untreated sample showed highest dielectric constant values compared to the treated ones. It was also reported that all used chemical treatments decreased the dielectric constant as well as the loss factor.

MAPP treated composites showed the least dielectric constant value. The researchers related that effect to the ability of the treatment that decreased the hydrophilic nature of jute yarns by reducing the moisture absorption. This in turn, caused a reduction in the orientational polarization that led to lower dielectric constant and loss factor values. Besides, the chemical treatment was able to reduce the number of voids and other irregularities that led to decrease the water absorption and hence the dielectric constant [52]. In another study, chemical modification utilizing a biodegradable zein coating for flax reinforced polypropylene (PP) composites was investigated by John et al. [53]. They studied the effect of chemical modification as well as fiber loading on the composites particularly, their thermo-physical and dielectric properties. Also the dielectric constant of the composites was found to be higher than that of polypropylene. It was also shown that the reinforcement of flax fillers in the polypropylene increased the relative dielectric permittivity. Besides, the composites of banana, hemp, and agave (both treated with maleic anhydride and untreated cases) with high density polyethylene (HDPE) resin were separately studied for both surface and volume resistivity with different fiber loading conditions [50]. It was reported that the surface resistivity decreases, whereas volume resistivity increases with an increase in fiber content in the composites. The effect of chemical treatment on the volume resistivity of different ratios of natural fibers/HDPE. In addition, it was also demonstrated that chemical modification of the fibers decreased both thermal conductivity and diffusivity due to the increase in the matrix interfacial adhesion. The effects of fiber treatment, fiber size and fiber loading on the physical and dielectric properties of oil palm fiber/linear low density polyethylene compression molded composites were investigated. They were able to predict both the density and the dielectric constant of the fiber and composite utilizing different models. They found the dielectric constant of the oil palm fibers to be in the range of 7.76–8.31 whereas, that of the composite was in the range of 3.22–6.73. It was also reported that Alkali treatment was able to reduce the first degradation temperature of the composite to 297.1°C. In addition, jute fabric/polypropylene composites treated with red dye solutions (0.1–1%, w/w) for different soaking times were investigated regarding the dielectric properties [53]. Alkali, permanganate, and stearic acid treatments were used in the study. The dielectric property of the composites was almost the same for all the chemical treatments but significantly increased with fiber
loading. The maximum value of dielectric constant was found to be 2.56 at 25% fiber loading. Besides, the effect of addition of porous additives on dielectric constant of sisal/polypropylene was investigated [54].

5. Various Natural fibres and its properties

Natural fibres are now considered as a serious alternative to glass fibres for use in composite materials as reinforcing agents. The advantages of natural fibres over glass fibres are their low cost, low density, high strength-to-weight ratio, resistance to breakage during processing, low energy content and recyclability [55]. Since they are waste, the utilization of natural fibres as reinforcement for polyester composite is a best eco-reusing technique [56]. Natural fibres can be divided into two groups: natural fibres, which are available in a fibre form, and fibres with a natural origin that are artificially produced from natural raw materials. Currently, glass fibre is the typical reinforcing material for polymer composites. Carbon fibre is used when there are more specialised and greater requirements (e.g., space technology, the aircraft industry, military applications and sports). However, carbon fibre’s production costs are one order of magnitude greater than those of glass fibre, and adhesion between carbon fibres and the matrix is also more difficult to achieve [57]. Natural fibres like flax, sisal, coir, hemp, etc. are becoming more popular because it has satisfactory strength properties along with a relatively low price and good biodegradability. A disadvantage of these fibres is that the consistency of the fibres cannot be guaranteed; they are sensitive to the moisture content of the environment, and they do not adhere well to a polymer matrix under moist conditions [58]. Considering that the fibre market is very competitive and that the economic and environmental requirements imposed on plastic structural reinforcing elements are increasing, the applicability of newer fibres is being examined in leading research institutes throughout the world. Basalt fibres which extracted from common volcanic rock could be a good option for reinforcing with polymer matrices [59]. Its chemical composition is closely similar to glass; its basic components are SiO2, Al2O3, CaO, MgO, K2O, Na2O, Fe2O3 and FeO [60]. Thus, over the last few years, intensive research has begun on the applicability of basalt fibre as a reinforcing material for polymers. Its melting temperature ranges between 1350 and 1700 °C. When cooled slowly, basalt solidifies as a partially crystalline structure. Basalt fibres can be used from 200 to 600 °C without any significant loss of mechanical properties [61-62]. To enhance fracture toughness, basalt fibres were introduced into concrete composites [63]. Author investigated the durability and mechanical properties of basalt fibres strengthening structural concrete [64]. Studies on the use of basalt fibres as reinforcements of polymer composites have focused mainly on polypropylene and epoxy resin matrix composites [65-70]. The polyester resin could be used for reinforcement due to its advantages like cost effective, easiness in processability, lower density, etc. Generally, surface modifications enhance the mechanical properties of fibres. In the past, there have been few studies on the surface modification of basalt fibre; however, the good chemical durability of basalt fibre has been mentioned in several articles. Most researches focus on the applicability, mechanical performance and interfacial properties of basalt fibre reinforced polymer composites but pay little attention to surface modifications.

6. Conclusions

All natural fibres can be considered environmentally friendly and non-hazardous materials. It is not a new material, but its applications are surely innovative in many industrial and economic fields, from building and construction to energy efficiency, from automotive to aeronautic, due to its good mechanical, chemical and thermal performances. Hence, natural fibres fibre has gained increasing attention as a reinforcing material especially compared to traditional glass fibres.

References

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