Effects of Coupling Agent on the Properties of Henequen Microfiber (NF) Filled High Density Polyethylene (HDPE) Composites

Pravin Gaikwad, Prakash Mahanwar

***Abstract*—**The main objective of incorporating natural fibers such as Henequen microfibers (NF) into the High Density Polyethylene (HDPE) polymer matrix is to reduce the cost and to enhance the mechanical as well as other properties. The Henequen microfibers were chopped manually to 5-7mm in length and added into the polymer matrix at the optimized concentration of 8 wt %. In order to facilitate the link between Henequen microfibers (NF) and HDPE matrix, coupling agent such as Glycidoxy (Epoxy) Functional Methoxy Silane (GPTS) at various concentrations from 0.1%, 0.3%, 0.5%, 0.7%, 0.9% and 1% by weight to the total fibers were added. The tensile strength of the composite increased marginally while % elongation at break of the composites decreased with increase in silane loading by wt %. Tensile modulus and stiffness observed increased at 0.9 wt % GPTS loading. Flexural as well as impact strength of the composite decreased with increase in GPTS loading by weight %. Dielectric strength of the composite also found increased marginally up to 0.5wt % silane loading and thereafter remained constant.

***Keywords*—**Henequen microfibers (NF), polymer composites, HDPE, coupling agent, GPTS.

I. Introduction

HIGH density polyethylene (HDPE) is one of the most widely used polymer, having numerous applications in making variety of bottles, different size of containers and various consumer goods because of its best quality stiffness, yield and impact strength as well as toughness and excellent environmental stress cracking resistance. In recent years, natural fibers have been exploited as a replacement over the conventional fibers such as glass, aramid and carbon as it is cheap and easily available in plenty. Natural fibers have good mechanical, high specific strength, harmless, ecofriendly and biodegradability characteristics. The tensile properties of natural fiber reinforced polymers (both thermoplastics and thermosets) are mainly affected by the interfacial adhesion between matrix and fibers.

Although the plant based natural fibers like jute, kenaf, flax and hemp have been widely used as reinforcement, leaf based natural fibers like sisal, banana and henequen have also been increasingly used. Out of them, henequen fibers are attracting more attention due to relatively low cost and density [1]-[6].

Henequen (Agave fourcroydes),which is a similar family with sisal, is a long, hard and strong fiber obtained from the long leaves of agave plants, which are native to the Yucatan, Mexico. These natural fibers have been used traditionally to make twines, ropes, carpets and cordages for a long period of time [7].

Silane coupling agents are predominantly used as mediators and as bonding agent between organic materials and inorganic materials. Hence as coupling agent silanes can improve the mechanical and electrical properties of materials in wet or dry conditions. Silane coupling agents are primarily used in reinforced plastics and electric cables composed of crosslinked polyethylene. Silane coupling agents also are being used in concrete, resins, sealants and adhesives. Other uses of silanes are in primers, paint technology, inks for printing and dyeing equipments etc. A variety of silanes have been applied as a coupling agent in the natural fiber reinforced composites to promote the interfacial adhesion and to improve the properties of the composites [8]. Z-6040 silane having chemical type as Glycidoxy (Epoxy) functional Methoxy Silane (GPTS) is a coupling agent used to improve adhesion of organic resins to inorganic surfaces. It is a low viscosity liquid and has properties like organic and inorganic compound for reaction, improves interfacial adhesion and increases strength of the composites. Lee and Cho reported the effect of natural fiber surface treatment on the interfacial and mechanical properties of Henequen/PP biocomposites [9]. Nilubol et al. investigated the effect of coupling agent on mechanical properties and morphology of $CaCO_3$ filled recycled High Density Polyethylene [10]. Also Liu et al. have studied effects of coupling agent and morphology on the impact strength of High Density polyethylene/$CaCO_3$ composites [11]. It has been analysed that the effects of coupling agent on mechanical and morphological behaviour of PP/HDPE blend with the two different $CaCO_3$ by [12]. Study of maleated PP as a coupling agent for recycled LDPE/Wood floor composites have been carried out recently by [13]. The effects of coupling agent on mechanical performance of Polyethylene composite comprising Wallastonite microfibers have been reported by [14]. The effect of coupling agent in Polyethylene-Wood fiber composites is reported by [15]. Chand and Dwivedi have studied the effects of coupling agent on abrasive wear

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behaviour of chopped jute fiber-reinforced PP composites [16]. Asumani et al. also investigated the effects of alkali-silane treatment on the tensile and flexural properties of short fiber non-woven Kenaf reinforced PP composites [17]. Mahanwar and Gaikwad reported the effect of coupling agent on the properties of short non-woven PET microfiber reinforced Polypropylene (PP) composites [18].

In this Investigation, we studied the effects of coupling agent with varying concentration to the weight % of total fibers on the properties of HDPE/Henequen microfibers composite which is earlier optimised at 8% weight of fibers to the weight of polymer matrix for the different silane compositions. The variation in weight % of Glycidoxy methoxy silane (GPTS) coupling agent on High Density/Henequen microfiber composite is totally the new approach of its kind which is to be analysed and optimised accordingly.

II.EXPERIMENTAL

A.Materials

The matrix polymer high density polyethylene (HDPE), injection molding grade with MFI 8gm/10min and 0.96 g/cc density was procured from Reliance Industries Ltd. Henequen microfibers (Agave fourcroydes) naturally found is a close relative of sisal plant obtained from village, Maharashtra, India. Glycidoxy (Epoxy) functional Methoxy Silane (GPTS) was obtained from Degussa, Germany. Isopropyl Alcohol was supplied by S.D.fine chem. Ltd.

B.Surface Treatment of Henequen Microfibers

Henequen fibers were chopped 5-7mm in length manually. The surface treatment of Henequen microfibers was achieved by soaking these fibers in Isopropyl Alcohol followed by silane coupling agent. The Isopropyl Alcohol was added as 100% (w/w) to the total fibers and Glycidoxy (Epoxy) functional Methoxy Silane (GPTS) was added as a coupling agent in varying concentration as 0.1, 0.3, 0.5, 0.7, 0.9 and 1% by weight of the total fibers. After the surface treatment these chopped natural microfibers were allowed to dry in an air circulating oven for 2-3 hours at steady temperature of 65^0C.

C.Preparation of The Polymer Composites

Each batch size constitutes as 8% Henequen microfibers in the total polymer matrix which is by experimentally found out as optimum value. These batches of HDPE/Henequen microfibers after alcohol-silane treatment were melt blended in corotating twin screw extruder (Model MP19 PC, M/S APV BAKER, UK) having L/D ratio of 25:1. The speed of the screw was maintained 60 rpm for all the compositions. The extrudates were quenched in a water tank at 20-25 ^{0}C and then palletized. The temperature profile for HDPE/Henequen microfibers composite was kept as Zone1-140^0C, Zone2-150^0C, Zone3-170^0C, Zone4- 180^0C and Die-190^0C.

D.Injection Moulding

The granules of the extrudates were predried in an air circulated oven at 80^0C for 4 hours and then injection molded in a microprocessor based Injection moulding machine (Boolani) fitted with a master mould containing the cavity for tensile, flexural and impact specimens. The processing parameters for injection molding were kept as Zone1: 150^0C, Zone2: 170^0C, Zone3: 190 ^{0}C for HDPE/Henequen microfiber composites.

III.CHARACTERIZATION

A.Mechanical Properties

The dumbbell shape tensile strength specimens were injection molded as per ASTM standards. . The tensile strength and elongation at break of the samples were evaluated according to the ASTM D638 M-91, using Universal Testing Machine LR50K [Lloyd Instrument Ltd., U. K.] The crosshead speed of 50mm/min was maintained for testing using a load cell of 50 KN. The results reported are average values of at least 3 test specimens. The flexural strength and flexural modulus was also measured using universal testing machine as mentioned above according to ASTM D790 M-92. Jaw speed of 2.8 mm/min was maintained for 3-point flexural strength and the span was kept as 60mm. All the reported values are average of at least three samples. The impact strength was determined as per ASTM D 256 using Avery Denison's pendulum type, Impact Strength Tester, [model 6709] with 2.7J striker. The results reported are the average values of at least 3 specimens.

B.Electrical Properties

The Dielectric Strength was determined using Zaran Instruments (India) with a 2mm thick composite disc as per ASTM D 149. The voltage was increased slowly and the voltage at which the current penetrated into the sample was noted. The results are average values of at least 3 specimens.

C.Rheological Properties

The melt Rheology of the HDPE/Henequen microfiber composites were studied using rotational rheometer (Physica MCR 101, Anton Paar, Germany), employing a parallel plate assembly, diameter 35mm, at 190^0C. The samples were predried before analysis. Melt viscosity [η (pa- s)] as a function of shear rate (1-100 s^{-1}) [y (1/s)] was recorded.

D.Morphological Properties

Scanning Electron Microscopy (SEM) was used to study the morphology of the composites. SEM studies of tensile test fractured samples were carried out using JSM-6380LA analytical scanning microscope of Joel make, Japan. The accelerated voltage used was 15KV. The samples were sputter-coated with platinum to increase conductivity of surface. The SEM images were recorded and interpreted accordingly.

IV. Results and Discussion

A. Tensile Properties

The results of tensile tests were shown in Table I. The variation of tensile strength of HDPE/Henequen microfiber (8 weight %) composite as a function of GPTS by weight % loading. Tensile strength of composite increased marginally with increase in the GPTS weight % concentration in the composite. The rate of increase of tensile strength is higher in the case of higher GPTS weight % concentration. The increase in the tensile strength may be due to the proper interfacial adhesion between microfibers and polymer matrix with the use of silane coupling agent.

Table I also depicts the variation of percentage elongation at break of the HDPE/Henequen microfiber composite. It is observed that the percentage elongation at break decrease with increasing concentration of wt % loading of GPTS. This is due to the interference of fibers in the mobility or deformability of the polymer matrix. This interference is produced due to the physical interaction and immobilization of the polymer matrix in presence of mechanical restraints which results in the reduction of elongation at break. It is attributed to the restriction of polymer chain movement.

Table II represents the variation of young`s modulus and stiffness of the HDPE/Henequen microfiber composite as a function of wt % concentration of coupling agent (GPTS). It is found that the stiffness and elastic modulus of the composite was increasing with the increase in wt% concentration of coupling agent. The increase in both the properties is significant up to 0.5wt% concentration of coupling agent and then both the properties decrease drastically. The stiffness and elastic modulus variation and their % increment up to 0.5wt% concentration of coupling agent clearly indicate that there is a proper dispersion of fibers into the matrix.

TABLE I
TENSILE PROPERTIES OF HIGH DENSITY POLYETHYLENE (HDPE)/ HENEQUEN (8 WEIGHT %) MICROFIBER COMPOSITE

Silane content (wt %)	Tensile Strength (MPa)	% Elongation (%)
0	22.50	43.2
0.1	22.54	40.64
0.3	22.73	38.45
0.5	22.89	37.55
0.7	22.91	35.21
0.9	22.95	31.94
1	22.80	26.03

TABLE II
TENSILE PROPERTIES OF HIGH DENSITY POLYETHYLENE (HDPE)/ HENEQUEN (8 WEIGHT %) MICROFIBER COMPOSITE

Silane content (wt %)	Young's Modulus (Mpa)	Stiffness (N/mm)
0	1728.53	414.83
0.1	1866.72	448.01
0.3	1890.23	437.65
0.5	1910.29	423.81
0.7	1980.56	373.24
0.9	1987.43	281.06
1	1877.53	275.45

B. Flexural Properties

Fig. 1 depicts the variation in flexural strength of the composite with weight % loading of coupling agent (GPTS).The flexural strength of the composite decreases with increase in wt% concentration of coupling agent. This can be attributed to the fact that there are no polar groups left on fibers or polymer side for interaction and may be the concentration of coupling agent may have reached to its maximum value. The interaction is varied because of the polarity and secondary forces available with coupling agent.

Fig. 2 represents the variation in flexural modulus of the composite with weight % loading of coupling agent (GPTS).The flexural modulus of the composite increases with increase in weight % concentration of coupling agent. This may be due to the strong interfacial adhesion between polymer matrix and fibers.

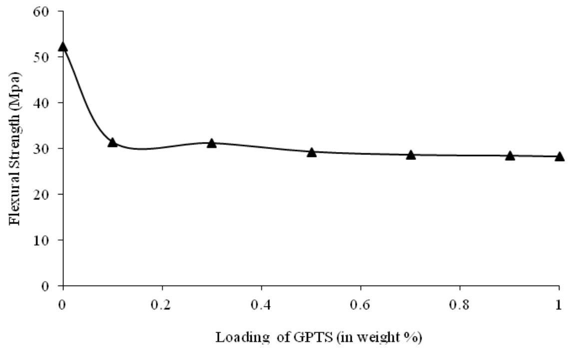

Fig. 1 Variation in flexural strength of HDPE/Henequen microfiber (8 weight %) composite with silane weight % loading

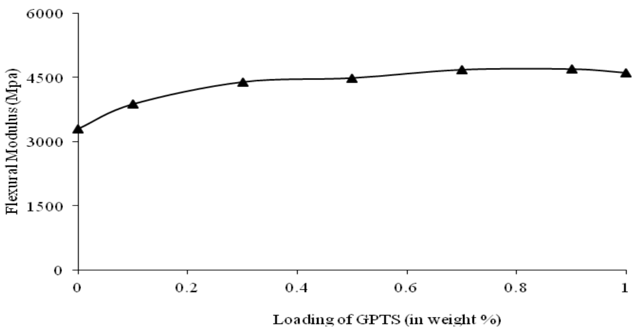

Fig. 2 Variation in flexural modulus of HDPE/Henequen microfiber (8 weight %) composite with silane weight % loading

C. *Impact Strength*

Fig. 3 depicts the variation in impact strength of the composite as a function of wt % loading of coupling agent (GPTS). It is found that with the increase in wt % loading of the coupling agent there is decrease in the impact strength of the composite drastically at 0.1wt% loading of coupling agent and thereafter increase marginally for 0.3 to 1wt% concentration. This is mainly due to the reduction in the elasticity of the composite due to fiber addition and thereby reducing the deformability of the virgin polymer. This is due to the fact that due to increase in the weight % loading of the coupling agent there is an increase in the crystallinity of the polymer matrix as molecules of the polymer gets attached to the fibers. This leads to the increase in the stiffness and decrease in the percentage elongation and impact strength of the composite.

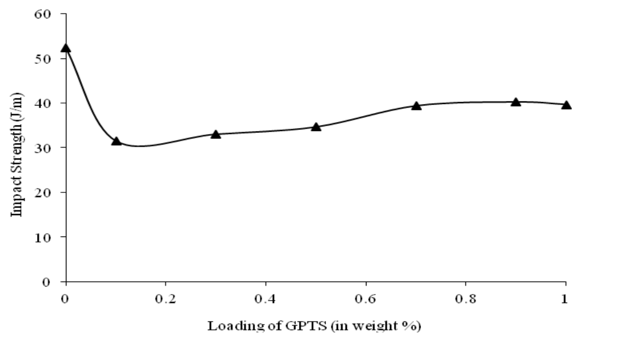

Fig. 3 Variation in impact strength of HDPE/Henequen microfiber (8 weight %) composite with silane weight % loading

D.Dielectric Strength

Fig. 4 represents the variation in the dielectric strength of the HDPE/Henequen microfiber composite with weight % loading of coupling agent. It is observed that the dielectric strength of the composite increase as there is an increase in weight % concentration of the coupling agent. The increase in dielectric strength is up to 0.5 weight % concentration of coupling agent. The concentration of the coupling agent may have reached the threshold value at 0.5wt% and therefore the dielectric strength of the natural fiber filled HDPE composites remain constant after 0.5 weight % concentration of coupling agent.

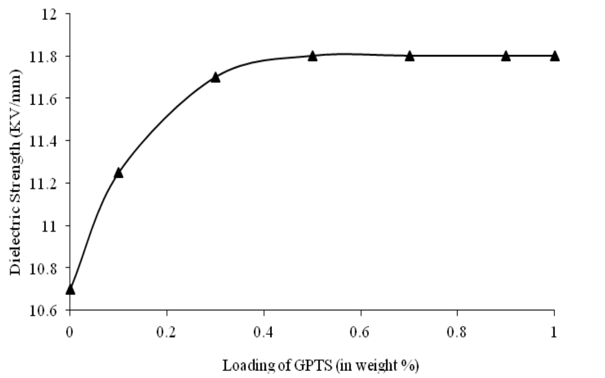

Fig. 4 Variation in dielectric strength of HDPE/Henequen microfiber (8 weight %) composite with silane weight % loading

E.Rheological Properties

Fig. 5 illustrates the variation of shear viscosities (in Pascal sec) with weight % loading of GPTS into HDPE/Henequen microfiber (NF) composites at 190^{0}C. The viscosity of the composite decreases as the weight % loading of the silane increases. Curve from the figure shows the shear viscosity of HDPE/Henequen microfiber (NF) with different weight % loading of silane is lower than without addition of coupling agent at low shear rates. Very high viscosity at low shear rates and relatively lower viscosity at high shear rates clearly indicates that onset of non- Newtonian flow behaviour for composite system.

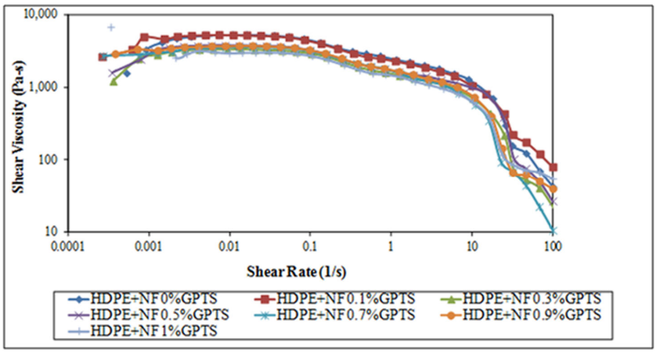

Fig. 5 Variation in shear viscosities of HDPE/Henequen microfiber (8 weight %) composite with silane weight % loading at 190^{o}C

F.Microstructure Characterizations

SEM is used to study the morphology of the composite. Fig. 6 shows the SEM micrograph of HDPE/Henequen microfiber (8 weight %) composite at 0.9 weight % concentration of silane coupling agent. It can be seen from SEM micrograph that at 0.9 weight % loading of silane into HDPE/Henequen microfiber composite evenly dispersion of fibers supports increase in mechanical and electrical properties of the composite.

Fig. 7 represents SEM micrographs of HDPE/Henequen microfiber 8 wt% compositions at 1 weight % concentration of coupling agent.

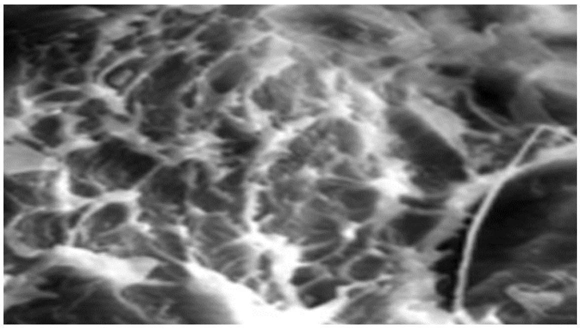

Fig. 6 Morphology of 0.9 weight% of silane loading into HDPE/Henequen microfiber compositions at 3000 x magnification

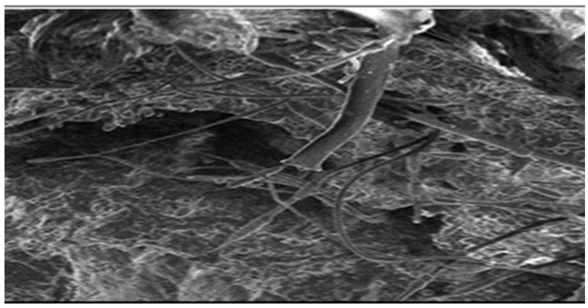

Fig. 7 Morphology of 1 weight % of silane loading into HDPE/Henequen microfiber compositions at 3000 x magnification

V. CONCLUSION

The principle conclusion obtained from the experimental results of HDPE/Henequen fiber at 8 wt% composite with the variation in wt% concentration of silane coupling agent (GPTS) are summarized as follows. Treatment with silane mainly results in increase of tensile modulus, flexural modulus and dielectric strength of the composites.

- The mechanical properties are enhanced when the content of silane coupling agent was increased from 0.1 to 1 weight % into the composite.
- An enhancement in tensile and flexural modulus of HDPE/Henequen microfiber composite was observed when fibers were surface treated with silane coupling agent.
- When microfibers were surface treated with 0.9 weight % coupling agent used for making composite, obtained optimum properties of HDPE/Henequen microfiber composite.
- Impact strength and percentage elongation at break of the HDPE/Henequen microfiber composites observed decreased at all silane weight % concentration.
- An enhancement in dielectric strength of HDPE/Henequen microfiber composites was observed when fibers were surface treated with silane coupling agent and found to be optimum for fibers treated with 0.9 weight % coupling agent.
- Morphological studies also revealed that at 0.9% by weight addition of silane treated microfibers concentration into the polymer matrix have uniform dispersion.
- The shear viscosity (η) of the composite and HDPE matrix, especially those for systems treated with silane, which was attributed to the interfacial adhesion enhancement.

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