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INFLUENCE OF ADDITIVES ON THE PHYSICO-MECHANICAL PROPERTIES OF SAWDUST- POLYETHYLENE (RECYCLED) COMPOSITES

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ABSTRACT

A number of additives are applied to composites in other to improve some of its end-use properties. The influence of additives on the physico-mechanical properties of sawdust- polyethylene (recycled) composites was studied. In this study, wood sawdust (250µm particle size)/pure water sachets (LDPE)/additives composites were prepared by compounding at temperature of $180^{o}C$ using Two Roll Mill Machine and Compression Molding Machineat $130^{o}C$ and 4pascal pressure for a period of 3 minutes. Generally, the physical and mechanical properties of the composites improved. The results on density of LDPE/Sawdust/Additives composites showed decrease in density with the addition of some additives (Calcium Stearate, Titanium dioxide and Sodium Silicate) while there was increased in density with addition of some other additives (Calcium Carbonate and MAH).The results obtained from water absorption after 30-day period showed increased in percentage of water absorption with increased in number of days in all the composites. Mechanical test of LDPE/Sawdust/Additives of all the composites improved. LDPE/Sawdust/MAH composite with density values of 1.3 g/cm^3 having good properties show good interaction and moderates interfacial bonding between the fillers and the matrix. The water absorption results obtained showed that LDPE/Sawdust/Calcium Carbonate composite has the lowest water imbibition (0.03%) compared to the control. Mechanical test of LDPE/Sawdust/MAH composite showed the highest tensile strength of 9.46MPa and Impact strength of 3.15 J/m^2. The composites produced showed good properties. It has low cost of production, low density, low processing requirements, renewability and eco-friendliness. The Moisture absorption of the composites is very low and tensile strength is high, therefore they are applicable for both indoor and outdoor usage.

Keywords*: Additive, Polyethylene (Recycled), Sawdust, Composite, Physico-mechanical, Compression molding.*

INTRODUCTION

Thermoplastic polymer matrix composites have gained commercial success in the semi structural and structural applications. Polyethylene (PE) is one of the most versatile and widely used thermoplastics in the world because of its excellent properties like toughness, near-zero moisture absorption, excellent chemical inertness, low coefficient of friction, ease of processing and unusual electrical properties. Natural fibers-reinforced composites have been used in countless consumer products, including cars, boats, skis, agricultural machinery, etc. during the last decades (Holbery *et al*., 2006). As the standard of living increases, the more prosperous consumers who are health conscious are increasingly turning to natural wood-based materials as construction materials for interior decoration and flooring. As the consumption of wood-based raw materials has increased, the need for wood substitutes has also grown (Yang, *et al.,* 2003).

Composite materials based on renewable resources such as natural fibers are environmentally friendly and low-cost alternatives to replace synthetic fibers like glass and carbon fibers (Khan *et al*., 2009). The use of this biomass as a filler in thermoplastics will allow the reduction of these scraps and waste. These bio-fillers exhibit a number of attractive advantages, including low cost, low density, low processing requirements, less abrasion during processing, renewability, eco-friendliness, and biodegradability (Demir *et al.,* 2006). Like other thermoplastic olefins, virgin and recycled polyethylene (PE) and polypropylene (PP) polymers have been used extensively in wood/polymer composites (WPCs) (Lin *et al.,* 2002), but very little investigations have been carried out to assess the effect of these additives on mechanical and physical properties of such composites. PE and PP have gained an important position among polyolefin because of their versatile and broad range of applications. However, the hydrophilic and aggregating nature of cellulose causes poor processability and inherent incompatibility with most hydrophobic polymers, such as PP. In the last 2 decades, many efforts have been made to improve the interfacial bonding strength between the polar wood fiber and the non-polar thermoplastic matrix (Haghighat *et al.,* 2005).

The incorporation of various additives and coupling agents (Mansour *et al.,* 2006) in these systems and the modification of the thermoplastics by grafting has helped to promote adhesion at the polymer–filler interface, improved the degree of filler dispersion, increased the fiber loading in the polymer, and improved the processability, moldability, and hence, the physico-mechanical properties. Additives are fairly important ingredients of WPCs that can be used for various purposes. The types of additives used in the WPC industry include lubricants and rheology control additives, coupling agents, stabilizers, fillers, density reduction additives, biocides, product aesthetics additives, flame retardants, and smoke suppressants. The effects of lubricant content on processing and property of wood flour/HDPE composites were explored (Adhikary, 2011). They found the apparent viscosity decreased by increasing the lubricant content. An optimal amount of wood fiber, HDPE, maleic anhydride polyethylene (MAPE), and lubricant can facilitate the processing by reducing viscosity and maintaining the mechanical properties and surface smoothness. Li and Wolcott studied the rheology of HDPE/maple composites after adding two distinct lubricants into the system: an ester and a zinc stearate (Li *et al.,* 2006). The ester-type lubricant enhanced the dispersion of maple particles and provided good external lubrication.

Biocides were employed to treat WPCs and their performance was evaluated (Simonsen *et al.,* 2005). The weight loss of the wood component in the WPCs caused by brown rot fungus was less compared to solid wood. The reduced decay of the wood is attributed to the plastic encapsulation effect. Na/Ca borate performed marginally better than zinc borate. Similarly, Dawson-Andoh, and Matuana, (2007) used two types of biocides (silver and zinc) at three concentrations to treat WPCs made from wood flour and HDPE (Dawson-Andoh, 2007). Overall, the zinc biocide controlled the fungal growth and discoloration effectively and it attained the best effect at 1 % (w/w) loading level. In contrast, the silver biocide did not inhibit the fungi or discoloration, which was deemed as inefficient for their study.

Ultraviolet (UV) light will cause WPCs to discolor and lose mechanical strength gradually. To overcome this durability issue, stabilizers like hindered amine light stabilizers (HALS) and ultraviolet absorbers (UVA) were applied to WPCs (Muasher, 2006). Diester HALS can stabilize the color of WPC by scavenging the free radicals generated by UV exposure. The higher the molecular weight of the diesters, the better stability it had in the composite. When combining benzotriazole UVA and diester HALS together, they reduced the discoloration of the composite synergistically. (Chaochanchaikul *et al.*, 2011) investigated three different UV stabilizers for wood/PVC composites. They found that UV stabilizers reduced the photodegradation of the WPC, however, did not appear to affect the mechanical properties. The same authors also studied the influences of thermal stabilizers on the structural and thermal properties of wood/PVC composites (Chaochanchaikul *et al.*, 2011).

The major challenge in compounding hydrophilic fillers and hydrophobic polymer matrices is poor interfacial bonding. It is therefore important to find a means of developing a covalent chemical bond between the matrix and the fillers using additives so as to improve the physical and mechanical properties of the composite. . Secondly, a number of additives are applied to composites in other to improve some of its end-use properties. However, very little investigations have been carried out to assess the effect of these additives on mechanical and physical properties of such composites. Thus, as the focus of this work, effect of some additives on physical and mechanical properties of such composites will be investigated.

In this sturdy, wood sawdust (250µm, 355µm and 500µm particle size)/pure water sachets (Recycled LDPE)/additives composites were prepared with wood sawdust as filler, pure water sachets (Recycled LDPE) as the matrix and additives are used to improve the physical and mechanical properties of the composites.

MATERIALS AND METHODS

Materials

Sawdust, Pure water Sachet (Recycled LDPE), Syringe, silicon oil, Disposable hand gloves, Foil paper, TiO_2, fine particle size limestone ($CaCO_3$), Copper stearate, Alumina trihydrate, Sodium Silicate.

Drying and grading of sawdust and Onion peel

Sawdust collected from Sabon-Gari, Zaria's Timber shed was air dried in the Weaving Laboratory of Polymer and Textile Engineering Department of A.B.U. Zaria, Nigeria for about five (5) days. The sawdust was later graded to 250 µm size.

Compounding

The used package water sachets for this research were separately compounded with the sawdust at temperature of 180°C using Two Roll Mill Machine (Reliable Rubber and Plastic Machinery Company, Model no 5183, North Bergen, New Jersey, USA). Five (5) different additives with the composite (5/20/75wt. %) i.e. Additive/Recycled LDPE/Sawdust was used, which act as stabilizer, antioxidant, coupling agent, thermal stabilizer or as filler.

Compression molding

The compression molding method involves pressing the composites at 130°C and 4pascal pressure for a period of 3minutes using a two steel plate mold lined with aluminum foil sheet lubricated with silicon oil for easy processing in Hydraulic Press (Compression Molding Machine, Wenzhou Zhiguang Shoes making machine company limited China).

Characterization of the composites

Density

The densities of the composite samples were determined according to ASTM D792-13. The composite samples were cut to a specific dimension (of 1x2 cm each) and immersed completely in the distilled water. The displacement of the water volume was observed and the density was calculated using the equation:

Density = mass of composite/volume of water displaced (g/cm^3).

Water absorption

Water absorption was conducted according to ASTM 2842. The composite samples were cut to a specific size (3x4cm) and weighed using electronic weighing balance.

The weighed samples were placed in a disposable plastic container and completely immersed in distilled water. The composites samples were left in the water for 24 hrs.

Thereafter, the samples were removed, cleaned with a soft cloth to remove surface moisture and reweighed. The same procedure was repeated for thirty (30) days and the percentage water absorption examined at 2 days intervals.

Scanning electron microscopy

Polymer composites are non-conductive, to make them conductive therefore, a sputter machine (model Q150R ES made by Quorum) was used with specification of 5nm gold to coat the surface of the sample before the micrographs were taken.

The coated samples were viewed through a navigation camera; proper adjustments were made to view the samples clearly before the machine was transferred to electronic mode. The viewing voltage was set using 10kV, the magnifications were increased and the sample morphology was stored in the electronic mode. Micro structures were obtained at various magnifications of 770X, 1000X and 1500X.

Mechanical properties

After the fabrication of the composites, the samples were conditioned for 24hrs and then subjected to the following mechanical tests: Tensile Strength, Hardness and Impact Strength. Each test was repeated five times for each sample of the composite and the average value was recorded.

Tensile strength

The test specimens in dumb-bell shape of the required standard dimensions according to ASTM D638 using "W" Monsanto Tensiometer and the machine were loaded. The values of the breaking load and elongation were taken accordingly.

Impact strength

Impact strength of notched samples was determined using the Cat Nr.412 Charpy Impact Testing Machine 15 joules capacity. The tests were conducted according to ASTM D-256.

Hardness

The Indentec Universal Hardness Testing Machine Model 8187.5 LKV "B" Rockwell HRF indentor (1/16" steel ball) with minor load 10 kg and major load 60 k was used in measuring the hardness using the shore scale according to ASTM D2240 at maximum time of 10 to 15 seconds.

FTIR

The samples sizes (20*20cm) were placed in the sample compartment of the Themo Nicolet No. Nexus 870 FTIR spectrometer. The infrared spectra of the composites were recorded and the spectrums were used for the analysis. The transmittance range of the scan was 4000 cm-1 to 650cm-1.

RESULTS AND DISCUSSION

Density

The results on density of LDPE/Sawdust/Additives composites in Fig. 1, show decrease in density with the addition of some additives (Calcium Stearate, Titanium dioxide and Sodium Silicate) while there was increased in density with addition of some other additives (Calcium Carbonate and MAH). However, the values of density of composites with the addition of additives obtained are compared to composite without additive. Low density value in composite with Calcium stearate was as a result of the fact that, it only served as an external lubricant in which it melted and plasticize the surface of the composite (Adhikary *et al.* 2010). Also low density value of composite with Titanium dioxide could be due to it degrading the composite and turn the composite colour white. Same also goes for composite with Sodium silicate, in which after reaction, increased porosity occurred due to volume shrinkage of the composite. Increased in density values of composites with Calcium carbonate and MAH could be due to their function in which they all served as reinforcement (filler). Thus, increased in weight of the composite occurred. General trend of composites with additives is in order of MAH > $CaCO_3$ >TiO_2 > $C_{36}H_{70}CaO_4$> Na_2SiO_3.

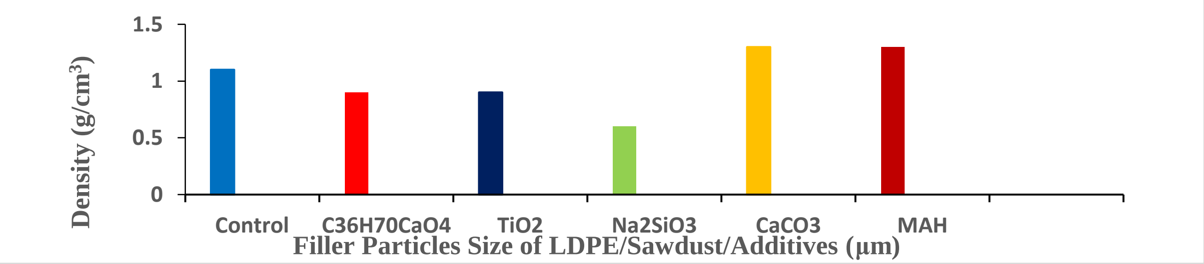

Fig. 1: Effect of additives on density of Composites

Water Absorption

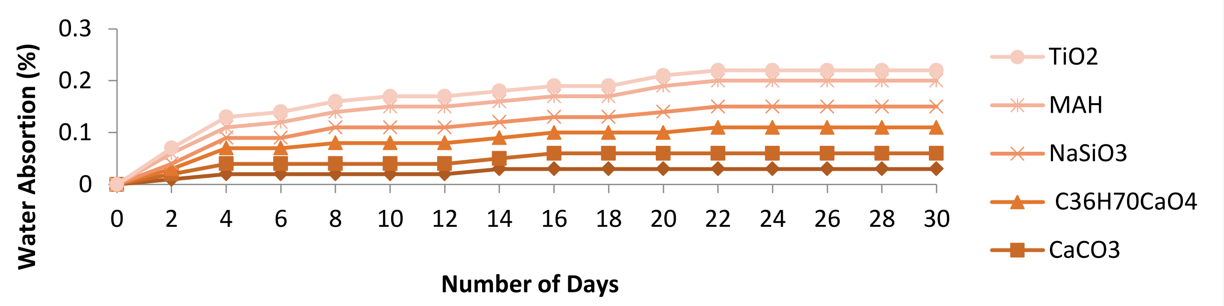

Fig. 2: Effect of time on water absorption of LDPE/Sawdust/Additives

From Fig. 2, it is shown that the LDPE/Sawdust/Additives composites showed higher water absorption compared to the control (composite without additive). However various additives have varying effects on water absorption. This could be attributed to the fact that, introducing additives which are mostly hydrophilic have increased the water intake of the LDPE/Sawdust/Additives composites because there are more polar groups in the polymer matrix (Kabir *et al*., 2011). Composite containing TiO_2 has the highest water absorption of 0.25 % followed by MAH with water absorption of 0.23 %, $NaSiO_3$ with water absorption of 0.18 % and Calcium Stearate ($C_{36}H_{70}CaO_4$) with water absorption of 0.12 % while composite containing $CaCO_3$has lesser water absorption of 0.05 % and 0.08 % respectively. This could be attributed to the nature of the chemicals (TiO_2, MAH, $NaSiO_3$ and Calcium Stearate ($C_{17}H_{70}CaO_4$) which easily absorbed moisture when exposed to moisture or water while $CaCO_3$ do not create room for void.The trend of water absorption of composites with additives is in order of TiO_2 $_>$MAH> $NaSiO_3$> $C_{36}H_{70}CaO_4$> $CaCO_3$.

Tensile strength

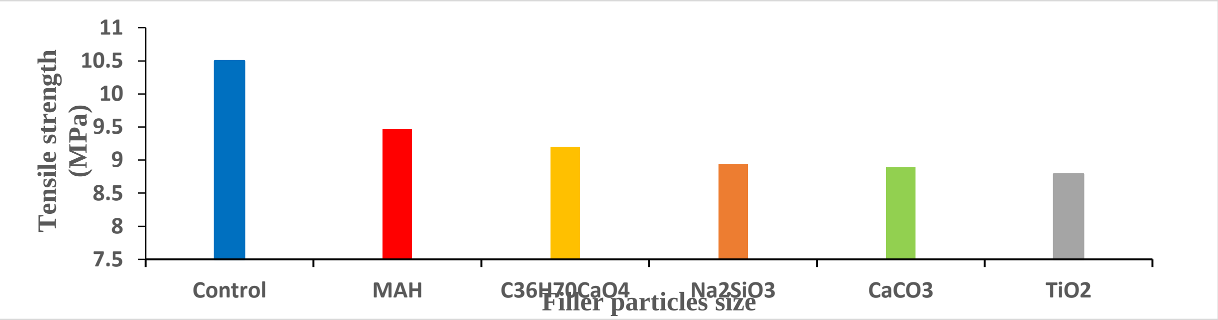

Fig. 3: Effect of additives on tensile strength of composites

From Fig. 3, it was observed that tensile strength increased with addition of additives to the composite when compared to composite without additives in MAH and Calcium Stearate from 9.11 MPa to 9.46 MPa (3.8%) and from 9.11 MPa to 9.2 MPa (1 %) respectively. Slight reduction of tensile strength was observed in the addition of sodium silicate, calcium carbonate and titanium dioxide when compared with the control (composite without additives). The increase in tensile strength in the composites with the addition of MAH and Calcium Stearate could be as a result of the fact that MAH (coupling component) and Calcium Stearate (thermal stabilizer) promote adhesion and evenly dispersion of the wood sawdust in the polymer (LDPE) matrix, thereby improving the tensile properties of the composite (Mansour *et al* 2006). Decrease in strength of the composites containing some other additives could be as a result of agglomeration of the filler which caused poor interfacial adhesion between the polymer and the filler when the additives react with the polymer. Strength trend of composites with additives is in order of MAH > $C_{17}H_{70}CaO_4$>Na_2SiO_3> $CaCO_3$ > TiO_2.

Modulus of Elasticity

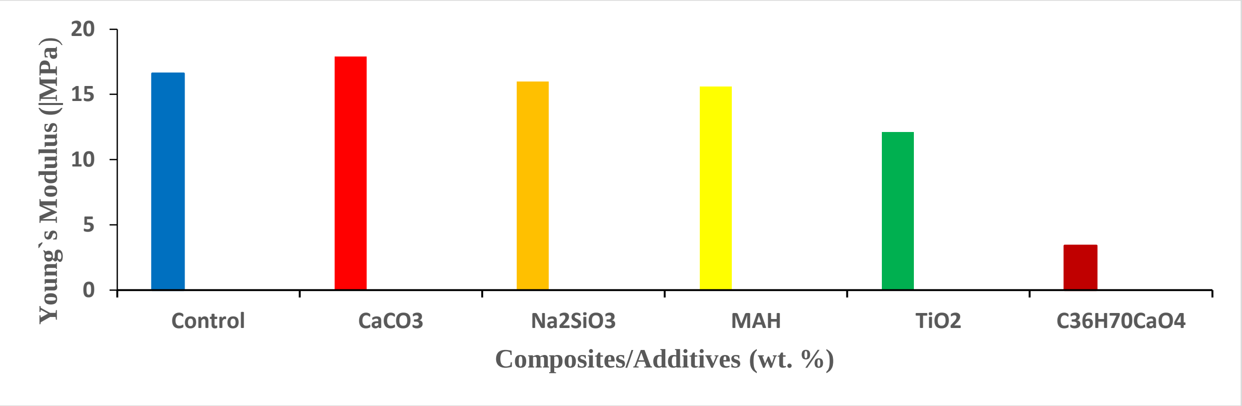

Fig 4: Effect of additives on modulus of composites

Fig. 4 shows varying values for Young’s modulus with highest value at composite with $CaCO_3$, 17.9 MPa. Young’s modulus of the composites with additives improved in composite with Calcium Carbonate compared to the composites without additives while composite with other additives have reduction in Young’s modulus. Increased in Modulus could be attributed to good filler distribution between the polymer and sawdust with the help of additives which can provide the effective stress transfer between the fiber and matrix thus increased in the strength of the composites while decrease in Young’s Modulus could be attributed to poor filler distribution in the matrix which result to poor interfacial bond between the filler and the polymer matrix in the presence of additives which caused the reduction in the strength of the composites. General trend of composites with additives is in order of $CaCO_3$ > Na_2SiO_3> MAH > TiO_2 > $C_{17}H_{70}CaO_4$.

Elongation at break.

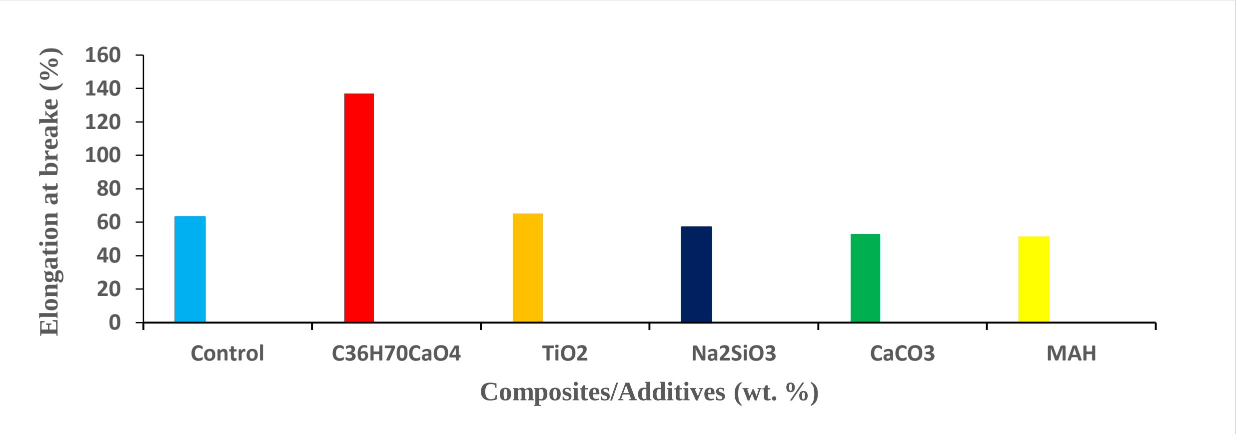

Fig 5: Effect of additives on elongation at break of composites

Fig. 5 shows that there was a large increase in elongation at break in composite with Calcium Stearate, from 52 % (for control) to 137 %. Also when the control is compared with the composite with Titanium dioxide, an increase of 25 % is observed. This is followed by decrease in elongation at break in composites with Sodium Silicate, Calcium Carbonate and MAH. The increase seen in composite with Calcium Stearate and Titanium dioxide can be attributed to better interlocking of the fillers with the matrix with the aid of additives present and the decrease could be to the formation of voids and the reaction of the additives with the polymer. The decrease in elongation at break in composites with additives (Sodium Silicate, Calcium Carbonate and MAH) may be attributed to the incapability of the composites to support the stress transfer from polymer matrix to the filler i.e. extension before breaking (Shuhadah and Supri, 2009).

The elongation trend of composites with additives is in order of C_{17} $H_{70}CaO_4$> TiO_2 > Na_2SiO_3 > $CaCO_3$ > MAH.

Impact strength test

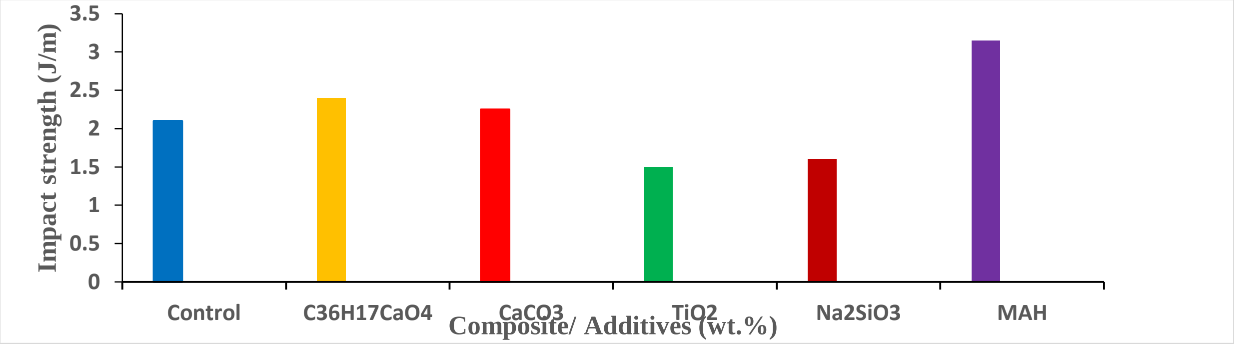

Fig 6: Effect of additives on impact strength of composites

Variation of impact strength with filler particles size for composites with additives is shown in Fig 6. Composites with MAH, Calcium Stearate and Calcium Carbonate show higher impact strength of 3.15 J/m^2, 2.4 J/m^2 and 2.25 J/m^2 respectively compared to the control while Composite with Sodium silicate and Titanium dioxide have lower Impact Strength of 1.9 J/m^2, 1.6 J/m^2 and 1.5 J/m^2 respectively compared to the control. Higher impact strength could be attributed to the fact that MAH, Calcium Stearate and Calcium Carbonate increased interfacial adhesion (compatibility) between the filler and the polymer matrix which improve the reduced impact strength caused by adding wood fibres into polymer matrices (Mansour *et al* 2006). Lower impact strength in composites with Sodium silicate and Titanium dioxide could be due to increase in brittleness of the composite as a result of imperfect adhesion between the components of the composite (Hristov *et al*., 2004). The trend of impact is in order of MAH>$C_{36}H_{70}CaO_4$>$CaCO_3$>Na_2SiO_3>TiO_2.

Hardness test

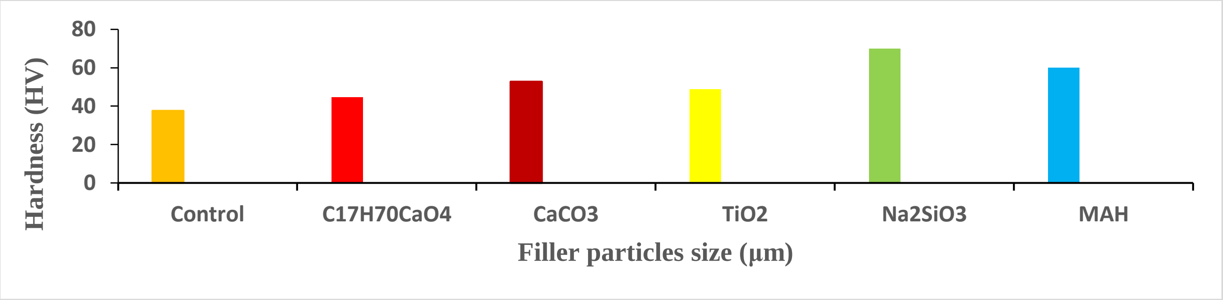

Fig. 7: Effect of additives on Hardness of composites.

Fig 7 indicates the hardness values of composites with additives. It was observed that composite with

Sodium silicate gave the highest hardness value of 70 HV followed by composite with MAH (60.1 HV), Calcium Carbonate (52.8 HV), Titanium dioxide (48.9 HV) and while composite with Calcium Stearate has the lowest hardness of (44.6 HV) compared to the Control (37.6 HV). Increased in hardness of the composite in the presence of Calcium carbonate could be attributed to good interfacial adhesion of the prepared composite which result to the increased in crystallinity of the composite. This is in line with the study of (Lyu S. G. et al, 2006) who reported that thermal and mechanical properties of composite containing Calcium carbonate in a LDPE composite improved the interfacial adhesion of the filler and polymer matrix. Same trend also applied to the other composites while poor interfacial adhesion in composite with Calcium Stearate could be the result of the reduction in hardness value. The trend of hardness of the composites, is in order of; Na_2SiO_3>MAH>$CaCO_3$>TiO_2>$C_{17}H_{70}CaO_4$.

FTIR

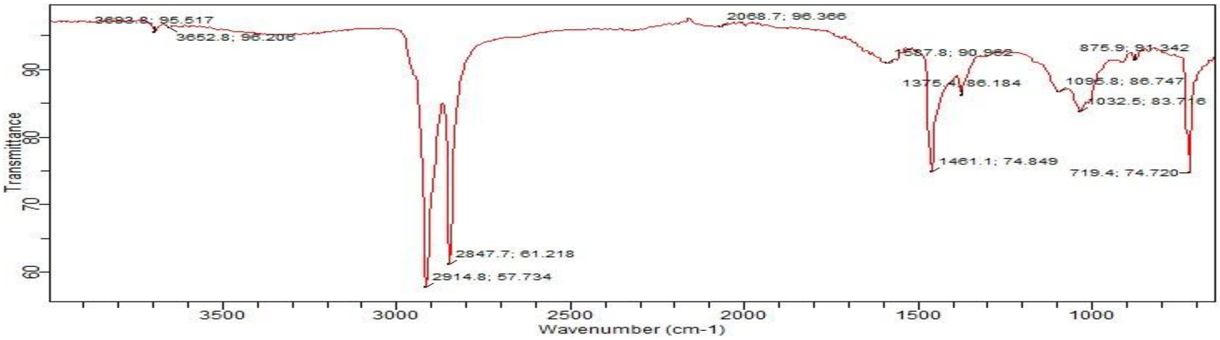

Fig 8: FTIR Spectroscopic of composite with MAH

The FTIR spectroscopic analysis of LDPE/Sawdust/MAH is shown in Fig 8. The spectrum clearly shows the absorption bands in 3693 cm^{-1} region (strong intensity), OH stretch is corresponding to cellulose, 1461 cm^{-1} (medium intensity), 2914 cm^{-1} region (medium) and 2847 cm^{-1} (medium intensity) which CH_2 deformation corresponding to methylene, CH stretch corresponding to methyl and $O\text{-}CH_3$ corresponding to aldehyde respectively. 719 cm^{-1} (weak intensity), CH_2 rocking corresponding to alkyl.The C=O stretching band of maleic anhydride was not shown in the spectrum, indicating no change in the group vibration according to whether the components are covalently linked or not (Guilherme *et al*., 1999).

CONCLUSION

This study shows the successful fabrication of LDPE/Wood sawdust/Additives composites. A covalent chemical bond between the matrix and the filler with the aid of additives was developed which improve the physical and mechanical and end-use properties of the composites. The results showed that density of the composites decreases considerably with increased in the filler. LDPE/Wood sawdust/MAH composites with values of 1.3 g/cm^3 having the better properties show good interaction and moderates interfacial bonding between the fillers and the matrix. The water absorption results obtained showed that LDPE/Sawdust/Calcium Carbonate composite has the lowest water imbibition (0.05%) compared to the control. Mechanical test of LDPE/Sawdust/MAH composite showed the highest tensile strength of 9.11MPa and Impact strength of 3.15 J/m^2, In general, the composites produced showed good properties. The Moisture absorption of the composites is very low, therefore they are applicable for both indoor and outdoor usage. The composites will be suitable for an innovative shading system to apply in the forefront of buildings, decking boards, office partition walls, indoor wardrobe for light weight clothing and shoe horns.

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