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Maleic Anhydride-graft-polyethylene Compatibilizer Effect on the Properties of Chrysophyllum Albidum Seed Powder Filled High Density Polyethylene

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Abstract Enhancement of chrysophyllum albidum seed powder (CASP) filled high density polyethylene (HDPE) using maleic anhydride - graft – polyethylene (MAPE) as a compatibilizer has been carried out. Using chrysophyllum albidum seed powder filler sieved to two particle sizes of 0.075 and 0.30μm and 0 to 30 wt %, high density polyethylene (HDPE) composites were prepared using an injection moulding machine. The mechanical properties; tensile strength, elongation at break, modulus of elasticity, and yield strength of the composites were determined using international accepted standard technigues. The mechanical properties decreased with increased filler (CASP) contents, and decrease in filler particle size. The addition of maleic anhydride graft polyethylene (MAPE) to the filler materials was found to improve the mechanical properties of HDPE composites. The specific gravity of high density polyethylene was greatly improved (>99 %) at high CASP particle size of 0.075μm. The hardness of HDPE composites was improved on addition of CASP filler. The addition of MAPE also improved the specific gravity of HDPE composites. The water absorption (24hrs) of the composites was found to increase with increased CASP content. The two particle sizes of the CASP investigated exhibited similar response to water absorption property. Generally the flame retardancy of the composites was found to increase with increased CASP content. Addition of MAPE to the composites was found to significantly improve the water absorption, and flame retardency even at low MAPE content of 0.035 wt. %. The addition of MAPE to high density polyethylene has highlighted the usefullness of chrysophyllum albidum seed powder as filler material in the plastic composite/industry.

Keywords: *chrysophyllum albidum, high density polyethylene, maleic anhydride-graft-polyethylene, composite*

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1. Introduction

The quest for polymeric materials with desired properties for specific applications has led to the relentless search for polymer blends. Since no one polymer has all the properties required in many application areas, polymers are frequently blended to improve their performance and processing characteristics. Natural fibers from different bio-renewable resources have attracted researchers all around the globe owing to their unique intrinsic properties such as biodegradability, availability, environmental friendly, flexibility, easy processing and good physico-mechanical properties. Natural fibers based materials are finding their applications in a number of fields ranging from automotive to biomedical [1]. Natural fibers have been frequently used as reinforcement component in polymers to add the specific properties in the final product. However, the growing global environmental concern, the high rate of depletion of petroleum resources, as well as new environmental regulations have forced the search for new fibre reinforced composite materials that are compatible with the environment. In this regard therefore, natural fibres have represented an environmentally friendly alternative to conventional reinforcing fillers such as talc, glass fibre, etc. The low cost, biodegradability, and high performance characteristics of natural fibres in polymer composites are of industrial and economic interest. Thus, a broad range of natural fibres are presently being utilized as the main structural components or as fillers in polymer composite [4]. The main limitation to the use of natural fibres in reinforcing polymers is the lower processing temperature permissible due to the possibility of fibre degradation and/or the possibility of volatile emissions that could affect composite properties, thus limiting the processing techniques of natural fibre components to about 200°C. Other drawbacks are the high moisture absorption, poor wettability, and general incompatibility with some polymeric matrices. Despite these limitations, the use of natural fibres in making composite is nowadays gaining importance.

2. Materials and Method

2.1. Materials Used

Polyethylene used in this study was obtained from Indorama Petrochemical Company LTD, Eleme, Rivers State, Nigeria. The polyethylene has a melt flow index (MFI) of 2.16 dg/min, and density of 0.946g/cm^{-3}. Chrysophyllum albidum, from which chrysophyllum albidum seed powder was extracted, was collected locally within Owerri Metropolis, Imo State, Nigeria. The chrysophyllum albidum seed was crushed and sieved to two particle sizes, namely 0.075 and 0.30µm respectively. Maleic anhydride – graft – polyethylene (MAPE) which was used as a compatibilizer in this study was a product of Sigma-Aldrich Cheme GmbH, Germany. It has a maleic anhydride content of 3.0 wt. %.

2.2. Preparation of High Density Polyethylene Composite

Table 1. Composition of HDPE Composites

S/N	HDPE Composite		HDPE Composite with MAPE		
	Filler content, wt. %	HDPE content, wt. %	HDPE content wt. %	Filler content, wt. %	MAPE content, wt. %
1	0.00	100.0	70	30	0.35
2	2.5	97.5	70	30	0.75
3	5.0	95.0	70	30	1.05
4	10.0	90.0	70	30	1.40
5	20.0	80.0	70	30	1.75
6	30.0	70.0	70	30	2.10

Two different sets of high density polyethylene composites were prepared. Firstly, the high density polyethylene/ crysophyllum albidum seed powder (HDPE/CASP) composites with filler particle sizes, 0.075, and 0.30µm were prepared at filler loadings, 2.5, 5.0. 10, 20, and 30 wt. %. Secondly, HDPE/CASP composites with a compatibilizer, maleic anhydride – graft – polyethylene (MAPE) were prepared with a fixed filler content of 30 wt. %. The MAPE used varied from 0.35 to 2.10 wt. % based on the weight of CASP. All the preparations were done using an injection moulding machine at a temperature of 150^{o}C. The resultant composites were produced as sheets of thickness, 2.18mm. The formulations used for the composite preparation are shown in Table 1.

3. Results and Discussions

3.1. Mechanical Properties of the Composites

The results of the mechanical properties of crysophyllum albidum seed powder (CASP) filled high density polyethylene have been determined and are represented in Figure 1 to Figure 10.

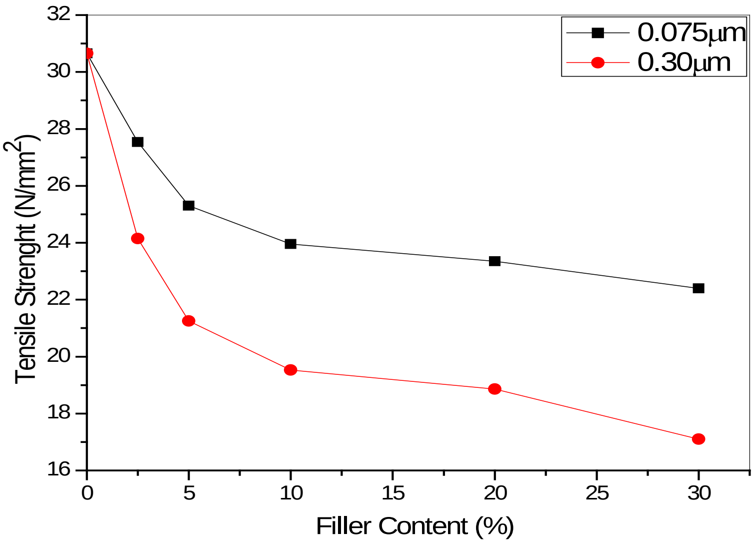

Figure 1. Plot of tensile strength versus filler content for CASP/HDPE composites at different filler particle sizes

The tensile strength of high density polyethylene composites decreased with increased filler contents at all filler particle sizes investigated (Figure 1). The decrease in tensile strength with increased filler content was probably due to a heterogeneous dispersion of the filler in HDPE matrix. The heterogeneous dispersion would cause significant stress concentration which could lead to formation of microvoids and quick development into crack during deformation [5]. The decrease of tensile strength with increased filler contents observed in this study was in agreement with the findings of Thakore et al, [10] who reported decreased tensile strength of linear low density polyethylene/starch blends with increased starch content. Similarly, Rozman et al [8] who worked with oil palm empty fruit bunch/polyethylene system reported decreased tensile strength with increased filler content.

Also from the above figure, there was an observed higher tensile strength of the CASP/HDPE composites with smaller particle sized filler at all filler content investigated. The better dispersion of the smaller sized filler, and consequently, the envisaged better filler - matrix interaction, may be responsible for the above observation.

The effects of the amount of maleic anhydride – graft – polyethylene on tensile strength of 30.0 wt. % of CASP/ HDPE composites are shown in Figure 2.

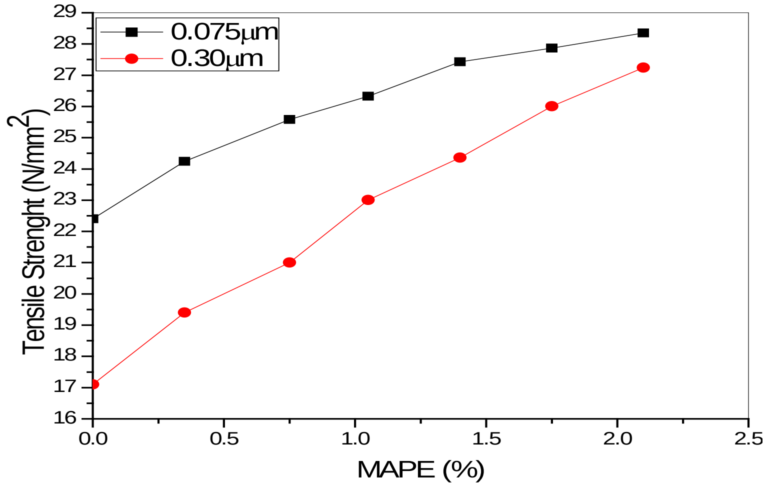

Figure 2. Plot of Tensile Strength versus Compatibilizer Content for CASP/HDPE Composite at different Filler Particle Sized

The addition of MAPE greatly improved the tensile strength of the composites. The resultant increases in the tensile strength of the composites as observed above was believed to be caused by maleic anhydride from MAPE molecule reacting with the hydroxyl group components of the crysophyllum albidum seed powder, leading to an esterification reaction between the filler and the matrix phase [2]. Furthermore, the long continuous chains in the MAPE molecules were compatible with the polymer matrix chains, via, physical entanglement. The combination of both the chemical and physical bonding led to improvement in tensile strength of the composite when compatibilizer was added. The increased tensile strength was in agreement with Chukwujike et al [1], who studied the mechanical properties of carbonized/uncarbonized cornhub powder filled natural rubber/acrylonitrile butadiene rubber bicomposite and reported increased mechanical properties of the composites with increased filler contents. The sharp increase in tensile strength therefore resulted from improved stress transfer from the matrix to the filler, via the compatibilizer. Further increase in the compatibilizer content beyond 0.75 wt. % for the CASP (0.075µm)/HDPE, and CASP (0.30µm)/HDPE composites had little effect on the strength of the composites. This may be that there was not much stress transfer from HDPE to the filler, irrespective of MAPE content above 0.75 wt. % of compatibilizer had been added at the two particle sizes of the filler used in this study.

The effects of filler content and particle size on elongation at break (EB) of high density polyethylene are shown in Figure 3. There was an observed decrease in EB of the composite with increased filler content for the two particle sizes of filler investigated.

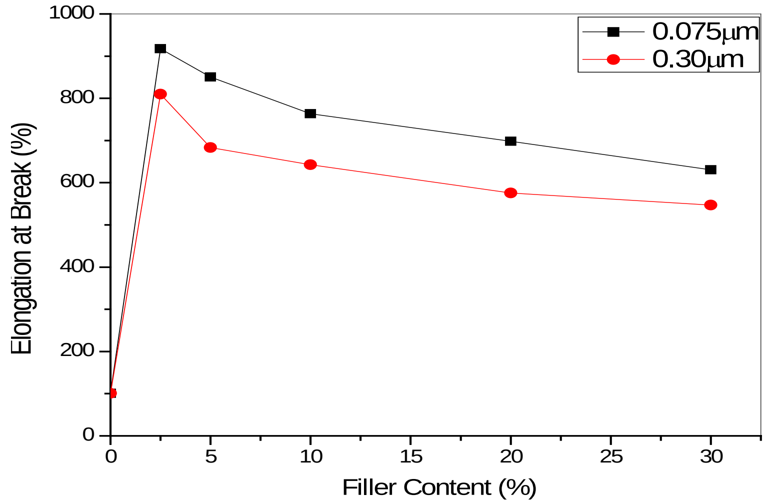

Figure 3. Plot of elongation at break versus filler content for CASP/HDPE composites at different filler particle Sizes

Generally, fillers can be considered as structural elements embedded in polymer matrix, and at the concentration of CASP considered (0 to 30 wt. %), the concentration might not be high enough to significantly restrain the polyethylene molecules. Consequently, highly localized strains might have occurred causing dewetting between the high density polyethylene matrix and the filler, leaving a matrix that is essentially brittle. Also, poor filler – matrix interaction or compatibility can be responsible for the observed poor ultimate performance. Such a reduction in EB of a composite with increased filler contents has been reported [8]. The increase in filler content greatly resulted in a reduction of deformability of a rigid interface between the filler and polyethylene matrix. The elongation at break (EB) of CASP/HDPE composites with MAPE is shown in Figure 4.

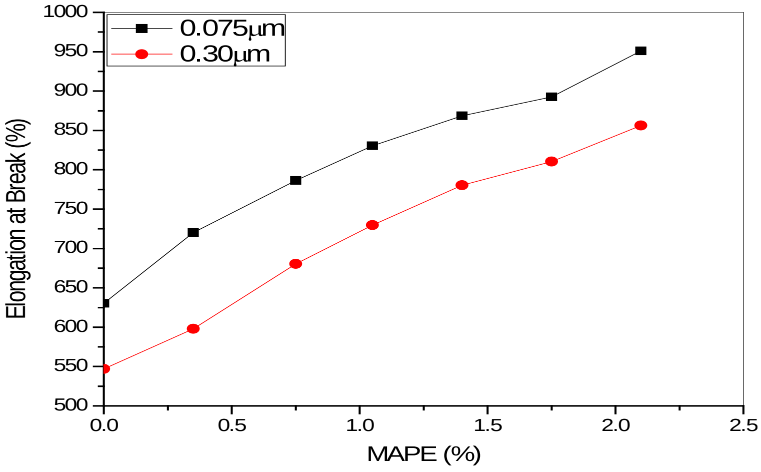

Figure 4. Plot of elongation at break versus compatibilizer content for CASP/HDPE composites at different filler particle Sizes

The elongation at break of the composites was significantly improved on addition of MAPE; improvement became more pronounced at high MAPE contents.

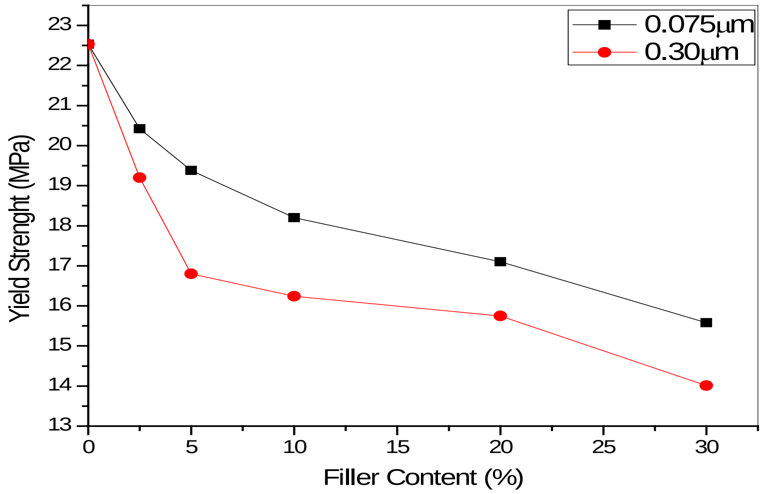

Figure 5. Plot of yield strength versus filler content for CASP/HDPE composites at different filler particle sizes

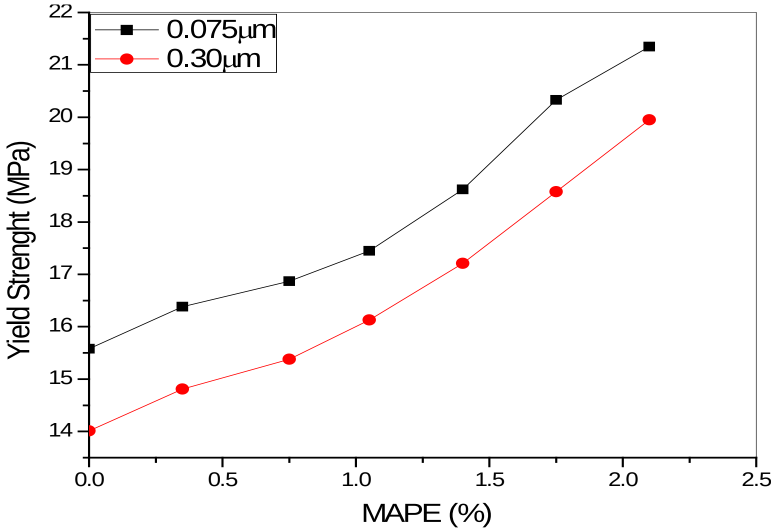

Figure 6. Plot of yield strength versus compatibilizer content for CASP/HDPE composites at different filler particle Sizes

The yield strength of the composites decreased with increased filler contents and particle sizes. Filler incorporation, as expected, caused material embrittlement, because the tougher high density polyethylene was replaced with bio filler [9]. The addition of MAPE at fixed CASP content greatly increased the yield strength of the composites (Figure 6).

The percentage changes in modulus 50, (M50) and 100 (M100) of HDPE on addition of CASP filler are illustrated in Figure 7 and Figure 8. The Figures revealed that the modulus of elasticity, M50 and M100 decreased with increased filler content at the two particle sizes of the filler investigated. The decrease of modulus of elasticity with increased filler content observed in this study was in agreement with the findings of Tavman [7] who worked on aluminium powder filled high density polyethylene and reported decreased modulus with increased aluminium powder content.

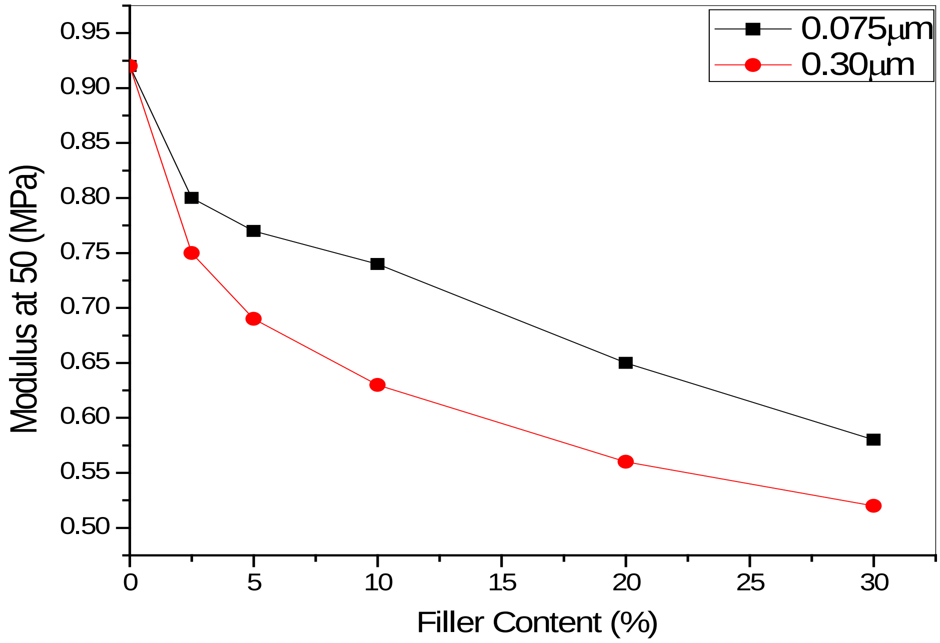

Figure 7. Plot of modulus M50 versus filler content for CASP/HDPE composites at different filler particle sizes

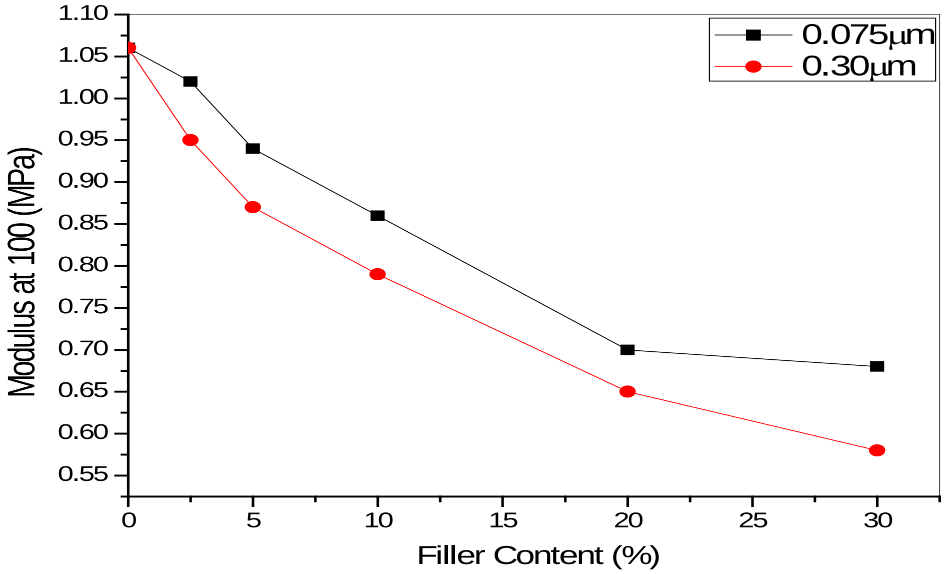

Figure 8. Plot of modulus M100 versus filler content for CASP/HDPE composites at different filler particle sizes

Such decreases in the modulus of elasticity could be attributed to the formation of cavities around filler particles during tensile stretching.

M100 was observed to be greater than that of M50 for the composites at any given filler content considered, irrespective of filler particle sizes. Similarly, both M50 and M100 of the composites were found to decrease with increased filler content for all the filler particle sizes considered. The changes in M50, and M100 of high density polyethylene composites in the presence of MAPE at fixed CASP content (30 wt. %) are illustrated in Figure 9 and Figure 10 respectively.

The modulus of elasticity of the composites was observed to increase with increased MAPE content. The Figures revealed that the addition of small amount of MAPE (0.35 wt. %) significantly improved the modulus of elasticity of HDPE composites. The increased modulus of elasticity of the composites in the presence of MAPE was associated with the improved compatibility, and interfacial adhesion between the matrix and filler particles.

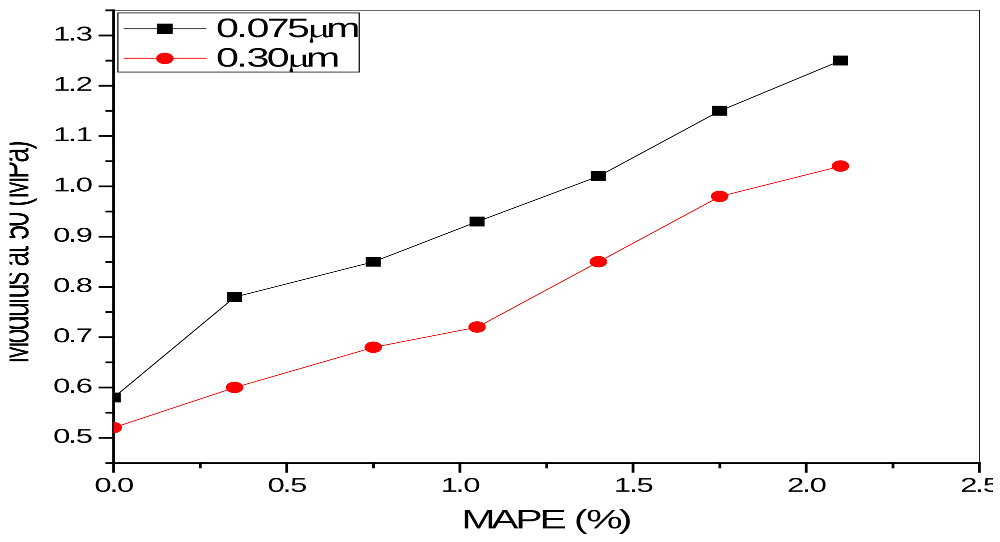

Figure 9. Plot of modulus M50 versus compatibilizer content for CASP/HDPE composites at different filler particle sizes

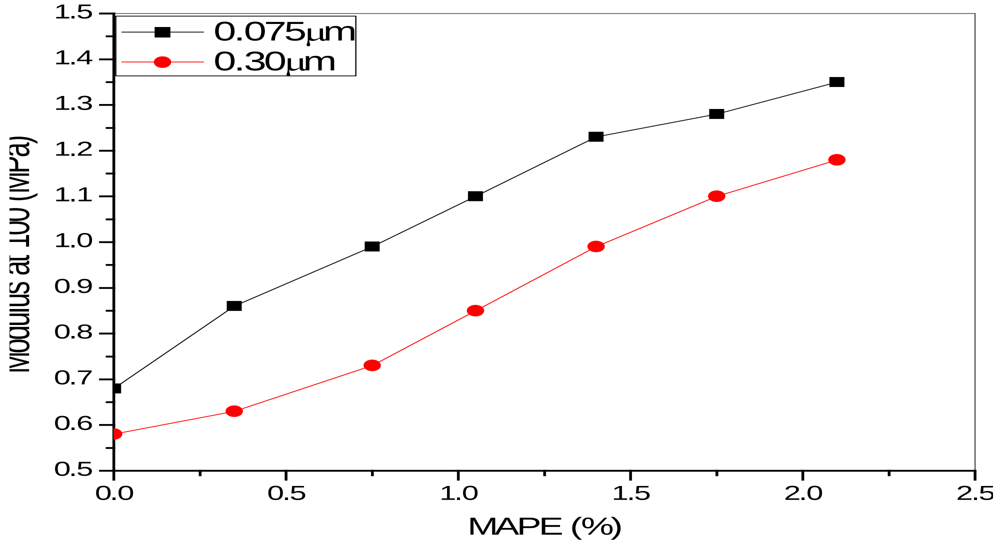

Figure 10. Plot of modulus M100 versus compatibilizer content for CASP/HDPE composites at different filler particle sizes

3.2. The End - Use Properties of the Composites

The experimental results on the end – use properties of chrysophyllum albidum seed powder (CASP) filled high density polyethylene have been determined and are shown in Table 2 and Table 3.

Table 2. Effect of Chrysophyllum Albidum Seed Powder (CASP) Content on the End-Use Properties of High Density Polyethylene Composite at different Filler Particle Sizes

Filler particle Size (µm)	Filler Content (%)	End - Use properties			
		Rockwell Hardness (MPa)	Specific Gravity (%)	Water Absorption (%)	Flame Propagation Rate (mm/sec)
0.075	0.00	66.00	1.013	0.039	1.04
	2.50	68.83	1.245	0.042	1.10
	5.00	71.15	1.502	0.043	1.15
	10.00	73.63	1.815	0.043	1.17
	20.00	78.68	2.023	0.045	1.17
	30.00	85.35	2.133	0.047	1.19
0.30	0.00	66.00	1.013	0.039	1.04
	2.50	67.51	1.115	0.043	1.15
	5.00	69.61	1.307	0.044	1.18
	10.00	72.83	1.568	0.045	1.19
	20.00	75.45	1.853	0.046	1.19
	30.00	76.23	2.020	0.048	1.19

Table 3. Effect of Maleic anhydride-Graft-Polyethylene (MAPE) on the End-Use Properties of High Density polyethylene Composites at different Filler Particle Sizes

Filler Particle Size (μm)	Filler Content (%)	MAPE (%)	End – Use Properties			
			Rockwell Hardness (MPa)	Specific Gravity (%)	Water Absorption (%)	Flame Propagation Rate (mm/sec)
0.075	30.00	0.00	85.35	2.133	0.047	1.19
	30.00	0.35	86.51	2.343	0.046	1.12
	30.00	0.75	89.80	2.562	0.046	1.00
	30.00	1.05	92.27	2.703	0.044	0.97
	30.00	1.40	96.05	2.832	0.043	0.97
	30.00	1.75	97.82	2.935	0.041	0.95
	30.00	2.10	102.32	2.987	0.040	0.91
0.30	30.00	0.00	80.23	2.020	0.048	1.19
	30.00	0.35	83.00	2.116	0.048	1.13
	30.00	0.75	84.56	2.205	0.047	1.00
	30.00	1.05	87.24	2.326	0.045	1.00
	30.00	1.40	90.63	2.501	0.043	0.98
	30.00	1.75	92.85	2.701	0.043	0.97
	30.00	2.10	95.33	2.815	0.041	0.93

The hardness of filled high density polyethylene increased with increased amount of filler. This result could indicate enhancement of abrasion and impact strengths of the composites. For reinforcing fillers, the composite becomes stiffer and harder with increasing filler content, which resulted to increased composite hardness. Generally, the hardness of the composites could be observed to decrease with increased filler particle sizes at any given filler content considered. The decrease in hardness of the composite with increased filler particle size is attributed to decrease in the degree of polymer – filler interaction associated with larger filler particle size.

A general increase in the hardness of HDPE composites with increased MAPE content was observed. The addition of MAPE was believed to have greatly imparted greater compatibility between CASP and HDPE. This was reflected in the greater hardness shown by the compatibilized composites.

The specific gravity of the composites was observed to increase with increased filler content at any filler particle size considered. There was also a general increase in the specific gravity of the composites with increased filler particle size, an observation attributed to be likely because of greater and more uniform dispersion of the smaller sized filler in the polymer matrix.

A gradual increase in the specific gravity of the composites with increase in MAPE content at a fixed filler loading (30 wt. %) was observed in this study.

Generally, the specific gravity of CASP/HDPE composites obtained in this study was much higher than those of mineral filled thermoplastic systems. Thus, the specific gravity of a 50 % (w/w) kenaf - polypropylene composite is about 1.07, while that of a 40 % (w/w) glass – polyethylene composite is 1.23 [7]. The water absorption indices (24 hrs) of the HDPE were found to increase when CASP was incorporated into it. The increase in the water absorption of the composites is quite similar, irrespective of the filler particle sizes. The increase in water absorption by HDPE when CASP was incorporated into HDPE was to be expected. The CASP was reported to contain mostly carbohydrates (54.6 %), a polyhydroxy compound, and proteins (26.5 %) [6]. Due to the presence of hydroxyl and other polar groups in CASP, the water absorption index was high, and increased with increased CASP content in the HDPE composites. This was envisaged to has caused the weak interfacial adhesion between the fillers and the hydrophobic matrix, which resulted to debonding. It was also observed that the addition of MAPE decreased the amount of water absorbed by HDPE composites. It has been reported that the maleic anhydride present in the maleic anhydride – g – polyethylene provides polar interactions such as acid – base interactions which can covalently link to the hydroxyl groups (-OH) on the biofiller [7]. The formation of covalent linkage between the maleic anhydride and the – OH group on biofiller surfaces have been indicated through ESCA analysis by Gatenholm et al [3]. The combination of covalent linkages and / or acid – base interaction between the MAPE and the –OH groups on the fibre surface resulted in good filler surface – interface properties. The resultant good adhesion decreased the rate, and amount of water absorbed in the interphase region of the composites. It was however difficult to entirely eliminate the absorption of water (moisture) in the composites without using expensive surface barriers on the composite surface.

Introduction of CASP into HDPE increased the flame propagation rate of the composite. The inability of the filler (CASP) to retard the rate of burning of HDPE composites could be attributed to poor energy absorption by the filler. When MAPE (>0.75 wt. %), was added into the composite at a fixed filler loading (30.0 wt. %), the flame propagation rate of the composites greatly reduced, irrespective of filler particle sizes. The use of MAPE appeared to have increased the energy needed to initiate burning in the system, and which was observed to increase with increased MAPE content, hence decreased the flame propagation rate of HDPE composites.

Generally, the burning characteristics of the composites in the presence or absence of the compatibilizer appeared to be the same. At 0.75 wt. % MAPE content, the flame propagation rate of HDPE composites appeared to be the same irrespective of filler particle sizes. This similarity was also noticed in the absence of MAPE when 30 wt. % of CASP was incorporated into HDPE. In essence, the filler particle size did not have much influence on the burning of HDPE composites.

4. Conclusions

The mechanical properties of crysophyllum albidum seed powder (CASP) filled high density polyethylene were found to decrease with increased filler content, and particle sizes. These properties (tensile strength, elongation at break, modulus of elasticity, and yield strength) were however improved appreciably on addition of maleic anhydride – graft – polyethylene (MAPE).

The use of crysophyllum albidum seed powder has resulted in improved hardness and specific gravity of high density polyethylene, properties which were further increased on addition of maleic anhydride – graft – polyethylene.

The water absorption (24 hrs) and flame propagation rate properties of high density polyethylene which were hitherto increased on addition of CASP were decreased upon the addition of MAPE into the composites. The use of MAPE has resulted to good wetting of the filler by HDPE, and which resulted to good CASP/HDPE bonding. There are reasons to believe that by understanding the limitations and benefits of CASP filled HDPE, crysophyllum albidum seed powder filler is not likely to be ignored by the plastics /composite industry for use in automotive, building, appliance and other applications. Material cost savings due to the incorporation of the relatively low cost crysophyllum albidum seed powder filler, coupled with the advantage of being non – abrasive to the mixing and moulding equipment, are a few of the benefits. Due to the lower specific gravity of crysophyllum albidum seed powder filler (0.98 g/cm^3) as compared to about 2.5 for mineral – based systems, there would be a definite weight advantage for these composites which may have implications in the automotive and other transportation applications.

The use of this natural product as a source of raw material to the plastic industry can not only provide a renewable resource, but could also generate a non – food source of economic development for the farming rural areas.

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