

Original Article

Mechanical behaviors of kolanut filled low-density polyethylene composite

G. C. Onuegbu, O. O. Nnorom, S. N. Okonkwo*

Department of Polymer and Textile Engineering, Federal University of Technology, Owerri, Nigeria

ABSTRACT

The mechanism of polymer reinforcement by fillers is not yet fully understood and is the subject of many publications in the scientific literature. In general, the properties of filled polymers change with the dispersion state, geometrical shape, and surface quality of the filler particles as well as the particle size. In this work, the mechanical behaviors of kolanut (KN) filled low-density polyethylene composite were studied. Different quantities of polyethylene-grafted maleic anhydride coupling agent were added to aid the adhesion between the polymer and the filler. The composite samples were produced using an injection molding machine. The addition of KN as filler into low-density polyethylene at the two particle sizes (0.30–0.45 μm) led to a decrease in tensile strength but increased the energy at breakage, modulus, impact strength, hardness, and flexural strength of the composite. The incorporation of coupling agent led to the increase in tensile strength and tensile strain. However, these characteristics decreased with increase in coupling agent content. Energy at break, modulus, impact strength, hardness, and flexural strength were enhanced further as coupling agent was incorporated into the composites.

Keywords: Coupling agent, filler content, kolanut, low-density polyethylene, mechanical behaviors, particle size

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INTRODUCTION

Natural fillers are hydrophilic and this makes them incompatible with polymer matrices which are hydrophobic in nature, consequently, to improve the mechanical properties of natural filler-reinforced polymer composite, it has become necessary to increase fiber-matrix adhesion through the incorporation of coupling agents.

Moreover, to achieve desired dimensional stability, strength, and other requirements in the application, filler plays a key role in plastics industry. The performance of polymer composites depends on the filler dispersion, the interactions formed at the polymer-filler interaction and the filler properties. The dispersion of filler in a polymer resin is important to arrive at expected application performance. For this to be achieved, a coupling agent can be used. A coupling agent is a chemical which improves the adhesion between two phases in a composite material.^[1,2] Coupling agents can be organosilanes, unsaturated acids, organometallics, acid-functionalized

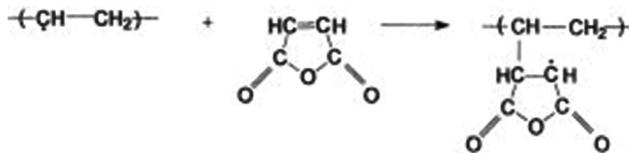
polymers, polymeric dispersants, and waxes (polyethylene and polypropylene).^[3]

Coupling agents can be used as dispersion enhancers in filling polypropylene/polyethylene master batch, flame retardant master batch, and degradable master batch because of its ability to interact with pigments, dye, and flame retardants. Coupling agents are dispersing agents that optimize the distribution of fillers in composites and during dispersing process, the coupling agents help to cover the newly formed surface of the aggregates and primary particle, thereby avoiding agglomeration of particles. Coupling agents can also be used as polarity enhancer. It does this by enhancing the polar group in the thermoplastic chain due to its excellent flexibility and compatibility.

The mechanism of the grafting reaction is shown in Equation 1 where the carbon-carbon unsaturation of the maleic anhydride group forms the bond with the polymer chain leaving the anhydride group free to react as an anhydride in

Address for correspondence: S. N. Okonkwo, Department of Polymer and Textile Engineering, Federal University of Technology, Owerri, Nigeria. E-mail: samdamian10@gmail.com

the newly formed polymer. The polar anhydride group on the polyethylene imparts the properties to the grafted polymer that makes these polymers good couplers for natural filler in polyethylene.



Structure of maleic anhydride grafted polyethylene (MAPE) Equation 1.

Composites are made from combinations of two or more materials present as separate phases and combined to form desired structures so as to take advantages of certain desirable properties of each component.^[4] The constituent of composites is the reinforcing agents which transfer stresses in the composite thereby improving the mechanical and physical performances and help to maintain fiber loading uniformity by reinforcing fibers along with flow as the matrix is moved on the mold. Another component of composites is the matrix which has many desirable physical, chemical intrinsic, or processing properties which can be improved by reinforcement to obtain certain engineering characteristics such as creep resistance, tear resistance, and tensile strength.

The example of polymer matrix is polyethylene which is classified mostly according to their branching and density. It can be high-density polyethylene, linear low-density polyethylene, and low-density polyethylene.

Low-density polyethylene (LDPE) has a density range of 0.910–0.40 g/cm³. It is not reactive at room temperature, but with strong oxidizing agents, it can be reactive at room temperature. It is translucent of opaque, flexible, and tough but breakable. It has weak intermolecular forces, its molecules are less tightly packed and less crystalline and are resistance to aldehydes, vegetable oils, ketones, dilute and concentrated acids, alcohols, bases, and esters.^[5]

The natural fillers are obtained from plants and animals. Example is kolanut (KN) which is a nut of kola tree with botanical name *Cola acuminata*. KN consists of caffeine (2–3.5%) and theobromine (1.0–2.5%), and it is highly toxic to the body. Excess intake of kola nut causes fever due to the presence of caffeine.^[6]

Polyolefin maleated polymers have been utilized as coupling agents in polyethylene and polypropylene composites and are reported in literature for improving the mechanical

properties of the composites as the result of strong interfacial bonding.^[7,8] Furthermore, Jianjian *et al.*^[9] used silane coupling agent as polarity enhancer in their study. Feiwen *et al.*^[10] incorporated coupling agents in the filling of polypropylene with bamboo.

In this study, effect of coupling agent on the mechanical properties of KN filled LDPE is studied.

MATERIALS AND METHODS

Materials

In this study, the LDPE used was gotten from Indorama Petro Chemical Company Limited Eleme, River State, Nigeria. The filler (KNs) used was locally sourced from Nkwo Mbaise Market, Ezinihitte Mbaise Local Government Area, Imo State, Nigeria. The KNs were washed, sun-dried, ground, and sieved to two particle sizes of 0.30–0.45 μm mesh sizes. MAPE, a product of Sigma-Aldrich Chemie GmbH, Germany, was used as coupling agent.

Composite Preparation

The KN filled LDPE at 0.30–0.45 μm particle sizes, at different KN contents (0–5 wt.%), and at calculated quantities of MAPE (0, 1, 1.5, and 2.5) was processed as previously discussed for 0.25 μm particle size LDPE composites.^[11]

Testing

The mechanical behaviors of LDPE/KN composites were obtained using standard procedures previously discussed.^[11]

RESULTS AND DISCUSSION

Mechanical Characteristics

The mechanical characteristics of LDPE/KN/coupling agent at 0.30–0.45 μm in the presence of different contents of the coupling agent are illustrated graphically in Tables 1 and 2 and in Figures 1-7.

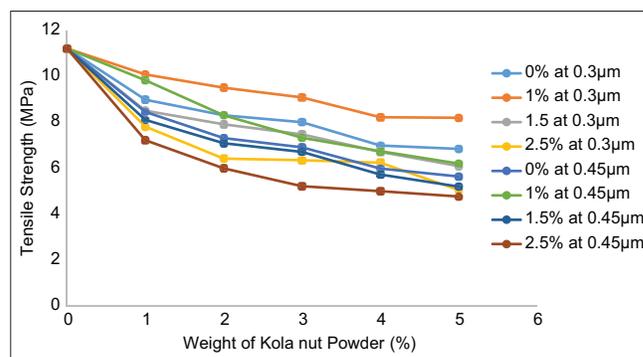


Figure 1: Graph of tensile strength of low-density polyethylene/kolanut/maleic anhydride grafted polyethylene composite at 0.30–0.45 μm particle sizes

Table 1: The effect of powder KN and coupling agent (MAPE) contents on the characteristics of LDPE composite at 0.30 μm particle size

Coupling agent content (%)	KN powder (%)	Tensile strength (MPa)	Strain at break (%)	Energy at break (J)	Tensile modulus (MPa)	Impact strength (J/m)	Hardness (BHN)	Flexural strength (MPa)
Virgin material	0	11.228	341.498	61.704	31.758	2.800	17.900	10.160
	1	9.000	198.678	60.567	44.235	2.900	20.000	10.610
	2	8.321	189.021	65.898	46.456	3.100	22.560	12.150
	3	8.021	180.678	68.234	48.323	3.200	24.670	15.670
	4	7.000	168.523	70.578	50.489	3.300	27.780	16.120
	5	6.852	158.824	74.618	51.678	3.400	30.470	18.210
1	1	10.102	200.235	70.345	45.436	4.000	20.800	12.560
	2	9.523	198.923	80.778	48.678	4.500	23.210	17.140
	3	9.100	188.273	82.693	50.234	4.800	26.600	20.570
	4	8.234	179.568	85.467	52.578	5.200	30.780	23.340
	5	8.210	165.235	87.245	54.478	5.300	32.670	25.480
1.5	1	8.523	164.56	80.352	48.373	4.300	22.150	20.670
	2	7.923	159.78	84.456	50.689	4.700	25.650	22.350
	3	7.489	150.32	86.579	53.845	5.000	28.580	25.790
	4	6.723	148.56	87.321	55.213	5.300	32.960	26.230
	5	6.105	141.78	90.253	60.313	5.600	36.230	28.770
2.5	1	7.824	151.4	81.679	50.124	4.700	23.768	23.130
	2	6.435	145.25	85.792	52.356	4.900	26.278	25.660
	3	6.362	138.45	87.679	55.679	5.200	30.678	28.230
	4	6.256	130.57	90.213	60.789	5.500	34.870	30.570
	5	5.0710	128.68	93.578	62.349	5.700	37.123	32.660

LDPE: Low-density polyethylene, KN: Kolanut

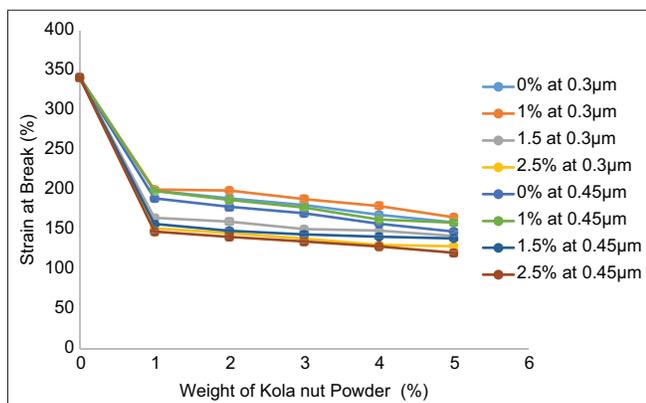


Figure 2: Graph of strain at break of low-density polyethylene/kolanut/maleic anhydride grafted polyethylene composite at 0.30–0.45 μm particle sizes

Tensile Strength

The effect of KN powder contents and coupling agent (MAPE) on the tensile strength at 0.30–0.45 μm particle sizes is illustrated in Tables 1 and 2 and Figure 1. It can be observed

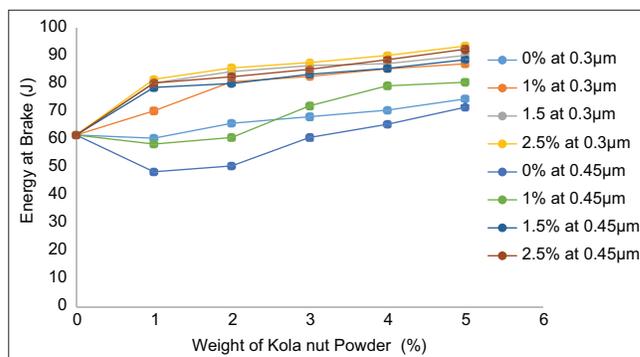


Figure 3: Graph of energy at break of low-density polyethylene/kolanut/maleic anhydride grafted polyethylene composite at 0.30–0.45 μm particle sizes

that the tensile strength of LDPE is 11.228 MPa. From the tables and Figure 1, the addition of coupling agent (MAPE) in the composite was observed to increase the tensile strength of PE/KN at 1 wt. % and later decreased as more coupling agents were incorporated into the composite for the two particle sizes studied. This is in agreement with Vedat^[12] and Vedat and

Table 2: The effect of powder KN and coupling agent (MAPE) contents on the characteristics of LDPE composite at 0.45 μm particle size

Coupling agent content (%)	KN powder (%)	Tensile strength (MPa)	Strain at break (%)	Energy at break (J)	Tensile modulus (MPa)	Impact strength (J/m)	Hardness (BHN)	Flexural strength (MPa)
Virgin material	0	11.228	341.498	61.704	31.758	2.800	17.900	10.160
0	1	8.456	189.234	48.479	40.479	2.810	18.230	9.970
	2	7.324	178.456	50.568	42.568	2.900	21.460	10.480
	3	6.923	170.346	60.812	46.327	3.100	25.78	13.550
	4	6.000	157.224	65.521	48.493	3.200	27.89	14.830
	5	5.654	147.340	71.642	50.235	3.300	32.17	16.780
1	1	9.845	198.482	58.463	44.583	3.800	18.52	12.230
	2	8.323	187.253	60.813	46.345	4.000	22.88	15.78
	3	7.345	177.673	72.126	48.973	4.200	26.093	18.56
	4	6.753	162.325	79.348	50.345	4.500	28.680	20.88
	5	6.213	158.538	80.579	52.467	4.900	32.56	22.75
1.5	1	8.123	156.87	78.745	46.123	4.100	18.930	18.46
	2	7.100	148.25	80.135	48.234	4.300	23.390	20.86
	3	6.725	143.78	83.478	50.878	4.500	26.230	21.63
	4	5.742	140.69	85.597	52.134	4.900	30.54	23.85
	5	5.213	138.74	88.672	54.250	5.200	32.98	25.83
2.5	1	7.234	147.23	80.345	48.235	4.200	20.345	20.86
	2	6.012	140.67	82.597	50.198	4.400	24.890	22.96
	3	5.230	134.67	85.235	51.235	4.800	26.780	26.00
	4	5.017	128.49	88.678	57.234	5.000	32.920	24.78
	5	4.780	120.57	92.482	60.459	5.200	35.470	30.56

LDPE: Low-density polyethylene, KN: Kolanut

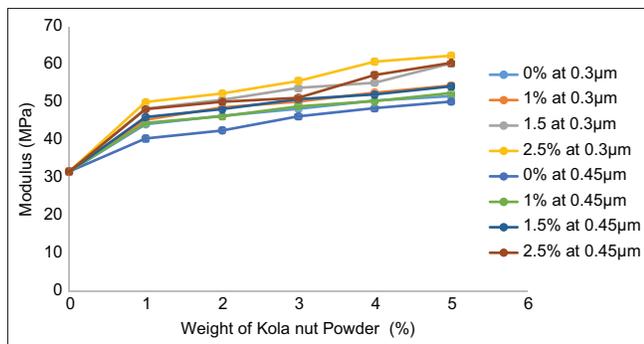


Figure 4: Graph of modulus of low-density polyethylene/kolanut/maleic anhydride grafted polyethylene composite at 0.30–0.45 μm particle sizes

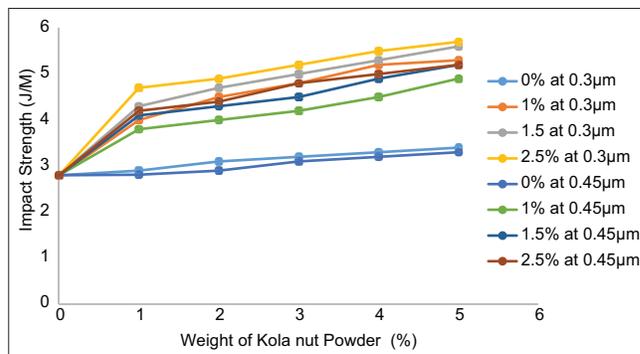


Figure 5: Graph of impact strength of low-density polyethylene/kolanut/maleic anhydride grafted polyethylene composite at 0.30–0.45 μm particle sizes

Mengelglu.^[13] The initial increase in tensile strength at 1 wt. % is due to the enhancement of the KN powder dispersion in the continuous LDPE phase, and the KN powder aggregates which disappeared with increase in the coupling agents, thereby leading to the homogeneity of the mixture. However, beyond

1 wt.%, the tensile strength decreased with increases in the coupling agent which is also in agreement with Reza *et al.*^[14] This suggests that there was no proper dispersion of the KN filler on the LDPE.

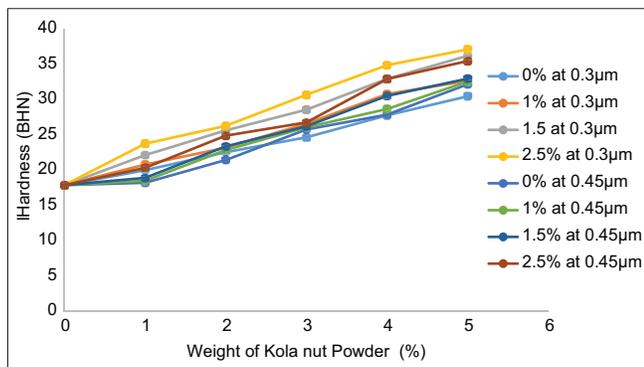


Figure 6: Graph of hardness of low-density polyethylene/kolanut/maleic anhydride grafted polyethylene composite at 0.30–0.45 μm particle sizes

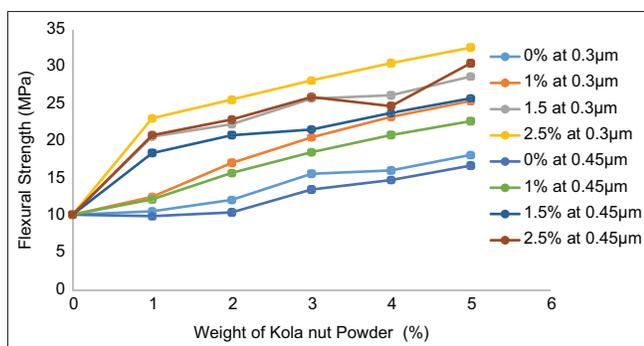


Figure 7: Graph of flexural strength of low-density polyethylene/kolanut/maleic anhydride grafted polyethylene composite at 0.30–0.45 μm particle sizes

The decrease in tensile strength of the composite with the incorporation of KN powder is in agreement with Arkles *et al.*^[2] and Vedat^[12] who worked on the effect of bamboo filler and coupling agent on the polypropylene composites and the effect of rice husk on the mechanical properties of polyethylene composites, respectively. However, it is contrary as seen in Onuegbu and Igwe^[15] who determined the effect of snail shell filler on the properties of polypropylene/snail shell composite and observed that the tensile strength increased with increases in filler content. Again, the tensile strength at 0.30 μm is higher than that at 0.45 μm particle size due to increase in surface area of 0.3 μm .

Strain at break

The strain at break of LDPE/KN/MAPE composites is shown in Tables 1 and 2 and Figure 2. The strain at break of unfilled low-density PE is 341.498%. The tables and the figure revealed that the strain at break of LDPE/KN composite decreased as more MAPE contents was incorporated into the composite. Increased filler loading in the LDPE matrix resulted in stiffening and hardening of the composite. This caused the reduction in resilience and toughness and further led to its low elongation at break. Similar results were also observed

by Eze *et al.*,^[16] and this decrease is attributed to the inability of the filler to support stress transfer from polymer filler to matrix^[17] as well as poor interfacial bonding. The strain at break of the LDPE/KN composites in the presence of MAPE is observed to decrease with increases in KN contents at the two particle sizes considered. Similar observation was made by Hemanth *et al.*^[18] and Khalid *et al.*^[19] who observed that there was decrease in the strain as the filler content increased. The reason for the decrease in strain at higher filler content is that it is more difficult for the polymer to penetrate through KN filler, thereby leading to poor transmission of stress across the filler-matrix interface. The strain at break for 0.45 μm is lower than that at 0.30 μm .

Energy at break

Data on energy at break of kolanut filled LDPE are given in Tables 1 and 2. These data are illustrated graphically in Figure 3. The energy at break of unfilled LDPE is 61.704 J. Figure 3 shows the increase in the energy at break of the composites with increases in coupling agent at the two filler particle sizes considered. This is due to strong adhesion between the matrix and the KN filler.

Figure 3 also shows the effect of the KN on the energy at break of LDPE composites at the two particle sizes studied. The figure and tables also show that there was a general increase in the energy at break of the LDPE composites with increase in the KN content. The energy at break of the composite made from 0.3 μm is higher than that of 0.45 μm .

Modulus

The data on the modulus of filled and unfilled LDPE are given and illustrated in Figure 4 and Tables 1 and 2. The modulus of unfilled LDPE is 31.75 Mpa. The tables and figure also show that the modulus of the composites was higher than the modulus of unfilled LDPE and increased with increases in MAPE content. This is as a result of interaction between the matrix and KN. This is in agreement with Obasi *et al.*^[20] Tables 1 and 2 and Figure 4 also show general increase in modulus with increases in KN powder content at the two particle sizes studied. The result obtained in this study is in agreement with the findings of Hamid *et al.*^[21] The modulus at 0.3 μm particle size is higher than that at 0.45 μm .

Impact strength

The impact strength of LDPE composites was obtained and given in Tables 1 and 2 and Figure 5. It is observed that the impact strength of unfilled polyethylene is 2.800 J/m. The data show that for each filler particle size, the increase in coupling agent increases the impact strength. Similar results were also observed by Armir *et al.*^[22] and Pankaj *et al.*^[23] Figure 5 also shows that the impact strength of the composites at the two particle sizes increased with increases in KN contents. This is due to uniform distribution of KN making the applied load to

penetrate into the matrix material and transfer around the filler. This is in agreement with Nwanonenyi *et al.*^[24] but contrary to Jia *et al.*^[25]

Hardness (Brinell)

Tables 1 and 2 and Figure 6 show the effect of coupling agent and KN filler contents on the hardness of filled LDPE. The hardness value of unfilled LDPE is 17.9 Mpa. Tables 1 and 2 and Figure 6 show that the hardness of all filled LDPE at a given filler particle size increased with increases in the coupling agent (MAPE) due to adhesion between the matrix and the filler. Similar results were observed by Onuegbu and Igwe^[15] and it is contrary to Arkles *et al.*^[2] The figure shows that the hardness of the composite at the two particle sizes investigated could be observed to increase with increases in the KN filler content. This result indicates the enhancement of the stress transfer by the matrix (LDPE). There have been reports which support increase in the hardness property of composite with increase in filler contents.^[16,17] This increase in hardness resulted in decrease in ductility of the composite formed, hence, ductility decreases with increase in filler content.

Furthermore, from Figure 6, it can be clearly seen that the hardness of the composites of 0.30 μm particle size of the filler is higher than that at 0.43 μm and this could be attributed to reduction in the degree of polymer-filler interaction at 0.45 μm particle size.

Flexural strength

The experimental data on the flexural strength of LDPE composites are illustrated in Tables 1 and 2 and Figure 7. The flexural strength of LDPE is 10.16 Mpa. Figure 7 shows that the addition of coupling agent promoted the flexural strength of LDPE/KN. This is due to strong adhesion between LDPE and KN and this is in agreement with Khalid *et al.*^[19]

At each particle size studied, the flexural strength of the composites increased with increases in KN contents. This is as a result of poor penetration of the load and transfer of the filler around the matrix, and this is in agreement with Jia *et al.*^[25]

CONCLUSION

The coupling agent (MAPE) and kolanut powder had effect on the mechanical characteristics of composite of LDPE studied. The MAPE improved the tensile strength (except at higher concentration of the MAPE), energy at break, modulus, and impact strength. However, MAPE decreased the strain at break as more MAPE contents were incorporated into the system. The use of kolanut powder at 0.3–0.45 μm increased the energy at break, modulus, impact strength, hardness, and flexural strength but it decreased the tensile strength and strain at break. Moreover, the particle size of the kolanut powder played a key role in the composites. The use of lower particle

size 0.30 μm improved the properties better than 0.45 μm in all the mechanical characteristics studied.

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