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THE EFFECTS OF LINEAR LOW-DENSITY POLYETHYLENE (LLDPE) ON THE MECHANICAL PROPERTIES OF HIGH-DENSITY POLYETHYLENE (HDPE) FILM BLENDS

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ABSTRACT
Polyethylene is a synthetic polymer that has been explored for various applications, such as films and packaging materials. However, studies involving HDPE/LLDPE film blends has received limited attention. The effect of LLDPE on the mechanical properties of HDPE film blends was investigated. Films of HDPE/LLDPE blends were produced by extrusion moulding using a Co-rotatory Twin-screw Extruder. The blends were characterized using Capillary Rheometer, Elmendorf-Type Tear Tester, MLANO/ITALIA Dart-Impact Tester and ILAO TIEH Instron Tensile Testing Machine, Model 1026. Results showed that, the Melt-Flow-Index (MFI) and tear (tensile) strength increased with increasing amounts of LLDPE, but the impact strength, yield strength and secant modulus decreased significantly.

KEYWORDS: Mechanical properties, Film blends, Extruder, Tensile strength, Dart impact.

INTRODUCTION

High density polyethylene (HDPE) is made by low pressure methods. In this method, pure ethylene is polymerized at a pressure of about 50 atmospheres and temperature between 60-200°C in the presence of Zeigler Natta catalyst system and supported oxide catalyst[1]. Such polymers are over 90% crystalline, sufficiently linear compared with low density type and density above $0.95g/Cm^3$. The low density polyethylene (LDPE) is made by high pressure methods. In this case, the pure ethylene is polymerized at a pressure between $1x10^3 – 3x10^3$ atmospheres, temperature of 250°C and using a 0.01% O_2 free radical initiator[1]. The crystallinity of such a polymer is low (50-60%), density is about $0.91 – 0.94g/Cm^3$ and the polymer is highly amorphous, highly branched due to chain transfer mechanism and readily dissolves in many organic solvents at about 100°C. New procedures for producing LDPE at lower pressures involving polymerization of alpha-olefin with ethylene usually from C_3 to C_8 carbon chain length has been evolved[1, 2]. These new LDPE have increased linearity, with a smaller amount of alpha olefins as co-monomers, density range of $0.918-0.940g/Cm^3$ and are referred to as linear low density polyethylene (LLDPE)

Blown films from high density homo-polymers tend to be very brittle because they lack tie molecules. Low density polyethylene however, is used mainly for the production of flexible materials such as films, while high density polyethylene is used for utility goods such as crates, seats and containers [3]. The LLDPE on the other hand, has improved physical properties quite superior to the conventional LDPE product especially with respect to their resistance to certain solvents. The use of highly active and stereo-specific catalysts, such as the Zeigler-Natta catalysts

has enabled product specifications such as linearity, specific density and melt-flow index. The linearity in the polymer structure imparts better tear and tensile properties[4], while the chain branching caused by the alpha-olefinic co-monomers, account for the inherent toughness of the LLDPE resin. These properties confer on LLDPE certain physical attributes which are generally more useful than those usually associated with the LDPE in several applications. It is tougher and has better tensile and impact properties, higher stress crack resistance and puncture energy. It also has better sealing characteristics and yields cheap end products by allowing up to 30% down-gauging when compared with LDPE [5, 6]. The heat seal and hot tack properties are excellent over broad sealing temperature ranges[7]. Linear low density polyethylene films are thus suitable for a host of applications, such as mulching, packaging of consumer goods, and industrial products such as fertilizers. Their outstanding tear strength in the transverse direction, TD, also makes LLDPE resins particularly suitable for stretch film applications.

The HDPE resins are used for the manufacture of heavy duty films for fertilizers, planting and merchandise bags/containers. Other applications include extruded netting, oriented tapes and monofilaments [8]. The mono- and multi-filaments of polyethylene, like polypropylene (PP), may serve a good purpose for making ropes, cordage, webbing and automobile seat covers. Bulk continuous filament is used in carpets, twines, brushes and fabrics. Most polymer films go into packaging applications, electrical insulation and constructional materials [9, 10]. The use properties of HDPE may be improved by one of four ways, namely: increasing the molecular weight, incorporating small

amounts of co-monomers such as butene, hexene, or octene, cross-linking and by blending with impact modifiers. Another less common method for toughening HDPE is achieved by adding some type of rubber. Though this method improves the "toughness" of the resin, particularly when it is converted into film, it tends to destroy some of the desirable properties of the polymer [11]. Addition of rubber decreases the modulus of the polymer and the barrier properties as the amount of rubber is increased. The same effect can be obtained by incorporating LDPE or LLDPE into the HDPE although these may improve some of the film properties[12].

Polypropylene has also been made tougher by blending with HDPE or LLDPE [13]. Ward [14] has also explored the potentials of blending PP with LDPE, LLDPE and HDPE in order to improve the mechanical properties of recycled polyolefins. The focus of this study therefore, is to determine the effects of LLDPE on the mechanical properties of HDPE blends. The results of this study will hopefully expand the applications of both polymers.

MATERIALS AND EXPERIMENTS

Materials: The linear-low-density polyethylene resin (grade F1U) and the high-density polyethylene resin (grade F8A) were obtained from the Plastic Technology Development Centre (PTDC), Eleme Petro-Chemical Company Limited, Port Harcourt, Rivers State, Nigeria and were used as supplied.

Preparation and Characterization of LLDPE and HDPE Blends

A sensitive electronic read-out weighing machine, Europe 1700 Model was used to measure a 100g of each blend portion of the LLDPE and HDPE pellets respectively. The blend ratios obtained by mixing LLDPE and HDPE resins are shown in Table 1.

Table 1: Blend Ratio (w/w)

Blend	1	2	3	4	5	6
HDPE (g)	100	90	70	50	30	10
LLDPE g)	0	10	30	50	70	90

Each of the blends was pulverized using a mechanical blender to ensure thorough mixing. The blended composition was charged into a Belton Extruder, London make, at the following operating conditions: *die-head temperature of 183 ± 1^0C, screw speed, 25 rpm and the motor load of 0.4 amps.* The blend composition was then passed through the die head orifice of the extruder which determines the thickness of the blown films. The extruded films had a blow-up-ratio (BUR) of 4.5:1, which signifies the ratio of the final diameter of the film (bubble diameter) to that of the die annulus.

Experimentals: *Tensile Properties*

The yield strength and secant modulus of the films, with specimen dimensions of 127mm x 12.7mm x 3mm, were determined using parallel-strip tensile test piece (ISO/DIS 527 Type D) on an Instron ILAO TIEH Model 1026 Tensile Testing Machine manufactured in France. The films were cut into parallel strip specimens and conditioned at a temperature of 23 ± 0.5^0C, relative humidity of $50 \pm 2\%$ and tested for tensile strength at a crosshead speed of 50 mm/min.

Melt Flow Index Test
The LLDPE and HDPE blends were fed into the cylindrical capillary rheometer with diameter of 0.65mm in which extrusion of the materials was accomplished at a working temperature of 190 ^0C, a pressure of 2.95 atm. and applied dead mass of 325g. The melt-flow index rate was quoted as a measure of the mass in grams of melted polymer extruded in 10 minutes through the capillary die.

Elmendorf-Type Tear Strength Test
Test pieces (200mm x 50mm) of the various films were cut out. A clean cut was made at one end of each specimen at a distance of 75mm (the cut was made central to the width of the film specimen). The two trouser legs were fixed in the jaws of the testing machine and a grip separation rate of 250 mm/min was maintained. The tear strength was obtained by dividing the force in the plateau region by the film thickness (3 mm).

Dart Impact Test
Izod and Charpy impact[15] on un-notched specimens was carried out on an Avery Denison Impact Tester using 2.7 J striker at a striking velocity of 3.46 m/s in accordance with ASTM D256. Specimen dimensions were (127mm x 12.7mm x 3mm). Tests were carried out on four samples. The test specimen was placed between the grips of the Izod tester and switched on. A sharp blow on the specimen breaks the test piece and the impact was recorded from the digital read out.

Results and Discussion:

Table II: Effects of LLDPE loads on: (a) MFI; (b) Tear Strength; (c) Impact Strength; (d) Yield Strength and (e) Secant Modulus of LLDPE/HDPE blends.

Parameters	Levels of LLDPE/ HDPE blend w/w (g)			
	0/100	10/90	30/70	50/50
MFI (g/10min)	0.09 ±0.01	0.14 ±0.02	0.30 ±0.02	0.40 ±0.04
Cv	11.11	14.29	6.67	10.00
Tear strength (N/m)	11.80 ±0.37	15.70 ±0.25	19.60 ±0.33	23.50 ±0.50
Cv	3.14	1.59	1.68	2.13
Impact strength (g/cm^2)	120 ±4.30	80 ±4.40	75 ±2.15	75 ±1.36
Cv	3.58	1.25	2.87	1.81
Yield trength(MPa)				
MD	28.80 ±0.43	27.40 ±0.51	23.30 ±0.42	18.50 ±0.40
Cv	1.49	1.86	1.80	2.16
TD	24.70 ±0.30	21.30 ±0.42	20.60 ±0.40	15.80 ±0.21
Cv	1.21	1.97	1.94	1.33
Modulus (MPa)				
MD	733 ±8.40	617 ±4.61	400 ± 5.17	350 ±7.86
Cv	1.15	0.75	1.29	2.25
TD	830 ±7.78	651 ±3.96	515 ±3.72	364 ±4.80
Cv	0.94	0.61	0.72	1.32

MD: Machine direction due to the axial-drawing of the film. Cv: Coefficient of variation

TD: Transverse direction due to the circumferential-drawing of the film.

Table II, shows the effects of varying loads of LLDPE on the mechanical properties of HDPE blend, namely melt flow index (MFI), Tear strength, tensile strength at yield, modulus of rigidity and impact strength. The effect of LLDPE loading on the melt flow index of HDPE blend is shown in Fig. 1. The result showed that with increasing LLDPE concentrations, the melt flow index of the composite increased. This may be attributed to the absence of branching in HDPE [16]. It is reasonable perhaps, to assign the difference in melt flow properties between the HDPE and that of LLDPE blend to the presence of short chain branching (SCB) in LLDPE. This is because the SCB tends to increase the entanglement at low shear rate (high elasticity), but at high shear rates the chain would disentangle, thus reducing the viscosity. Fig. 2 also shows that increasing the LLDPE loading increased the tear strength in the machine direction (MD) of the LLDPE/HDPE film. This is probably due to the linearity of

the linear-low-density polyethylene, which is believed to impart better tear strength[4]. The SCB in LLDPE, which is caused by the alpha olefinic co-monomers, accounted for the inherent toughness of the LLDPE –HDPE blend.

Figures 3, 4 and 5 showed significant decreases in impact strength, of 120 g/cm^2 for the control to 75 g/cm^2 at 50g LLDPE loading, yield strength and modulus of the film with increasing LLDPE concentrations in the blends. The decrease in impact strength is however unexpected and in contrast with earlier studies as linearity generally impart strength. Although the mechanical strength of a polymer film can be assessed from impact, tensile, and tear strength measurements, which are expected to be high, it is hardly possible to obtain high impact strength and high tear strength at the same time from a given polymer. It is also evident from Fig. 4 that the yield strength decreased from 28.8 MPa in the machine direction and 24.7 MPa in the transverse direction (TD) in pure HDPE to 18.5 and 15.8 MPa in the

machine direction and transverse direction respectively when 50 g of LLDPE resin was added. The decrease in the yield strength indicates a reinforcing action of the LLDPE on the HDPE resin. The observed decrease in the yield strength were understandable in line with the arguments of Eboatu et al [17] that a material is at a higher strength before the application of stress, and as stress is applied to it, the material slacks in tension, with tensile strength decreasing at yield point. As the LLDPE loading increased in the blend, the more or less rigid material became less rigid and therefore prone to creep.

The reinforcing action of the LLDPE on HDPE in the blend was also evident in the observation depicted in Fig. 5. This figure showed a decrease in modulus of rigidity (secant modulus) of the HDPE from 733 MPa in the machine direction and 830 MPa in the transverse direction at Og LLDPE to 350 and 364 MPa MD and TD respectively at 50g LLDPE. These observations were also in agreement with Ergoz et al, [18] and Brown[19], that disentanglement or rupture of tie-molecules was the dominant molecular mechanism in environmental stress cracking of polyethylene and in slow crack growth. The tie-molecules have also been identified as exhibiting similar mechanisms in impact and yield strengths. Thus, tie-molecules are important to all strength properties of polyethylene. Hence the increasing concentrations of LLDPE introduced tie-molecules into the polymer blend. Brown et al [20] have also reviewed slow crack growth in polyethylene and observed that the basic failure process in slow crack growth involves disentanglement of tie-molecules.

CONCLUSION

The incorporation of LLDPE into HDPE acted as a reinforcing additive .The melt flow index and the tear strength of the HDPE resin separately increased by 33.3%, hence a better processibility and utility of the rigid polymer. Also, the impact strength, yield strength and modulus of rigidity decreased significantly showing an inverse relationship with the tear strength.

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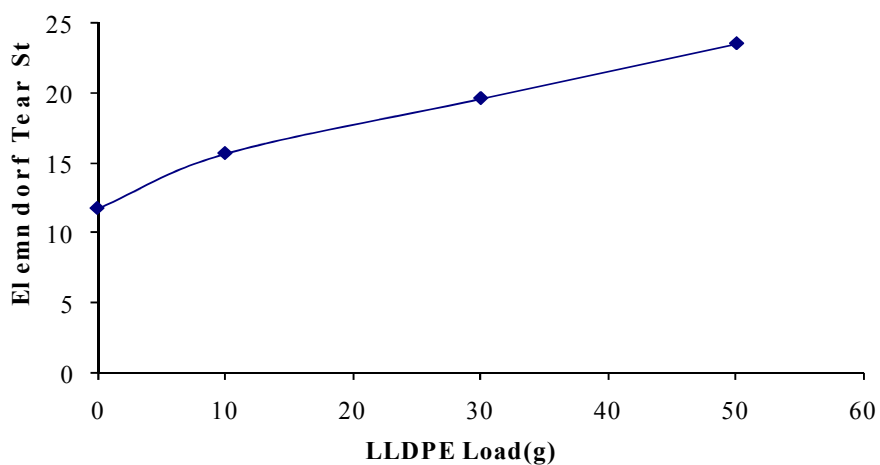
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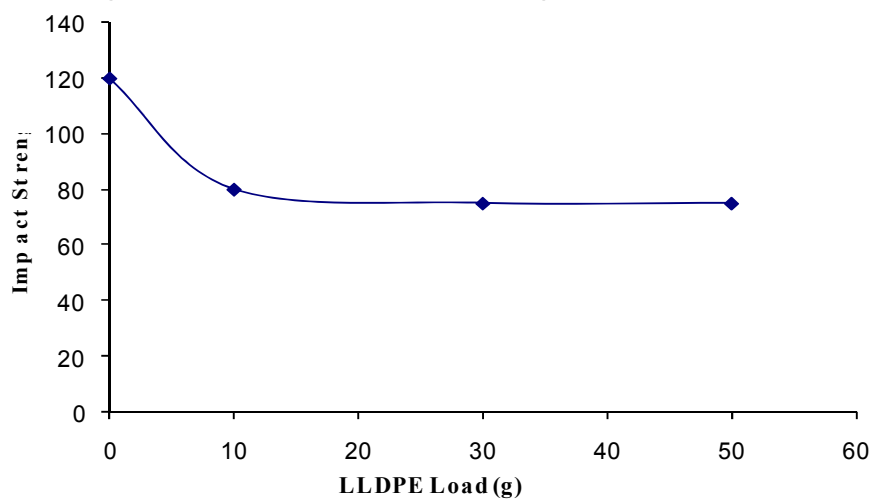
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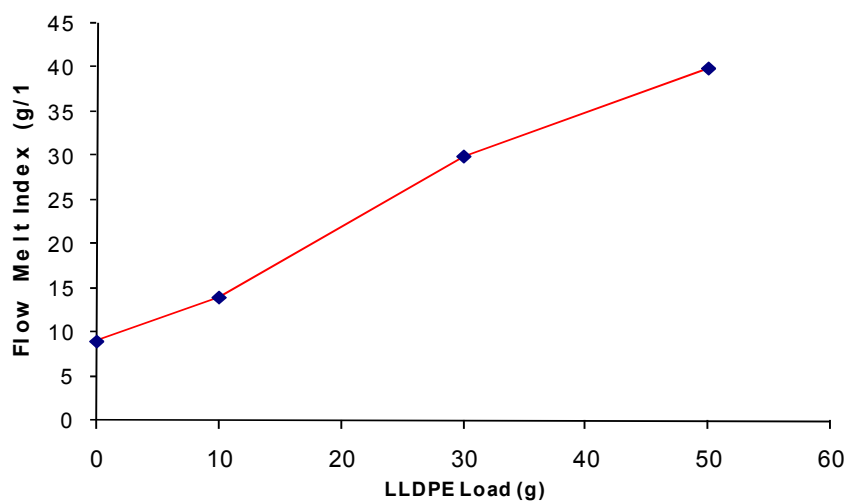
Fig.1: Effect of LLDPE on the Melt Flow Index of HDPE / LLDPE Blend.

Fig. 2: Effect of LLDPE on the Tear Stength of HDPE / LLDPE Blend.

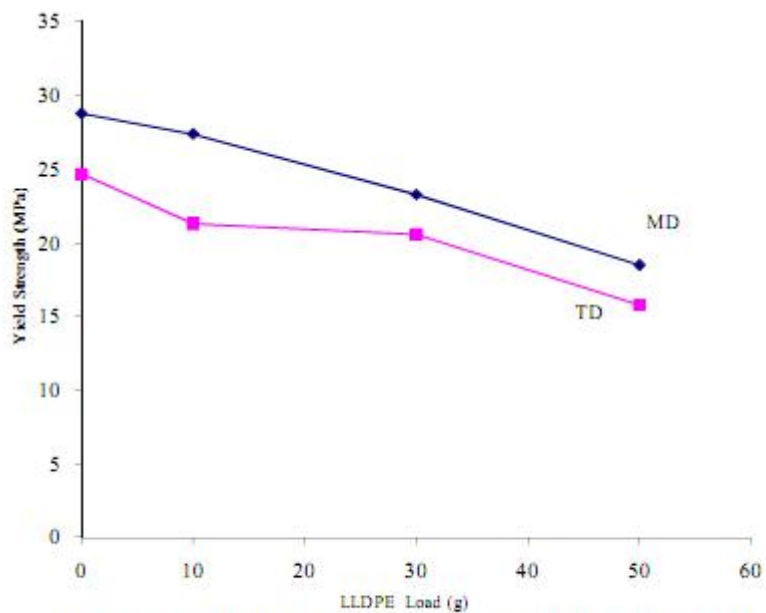**Fig. 3: Effect of LLDPE on the Impact strength of HDPE / LLDPE Blend**

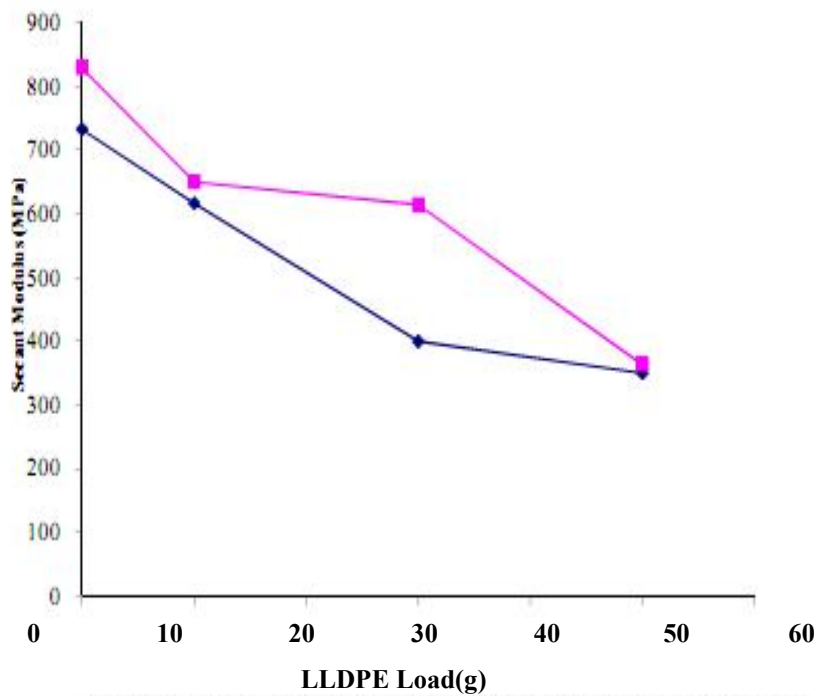Fig. 4: Effect of LLDPE on the Yield Strength of HDPE / LLDPE Blend.

Fig. 5: Effect of LLDPE on the Secant modulus of HDPE/LLDPE BLEND