International Journal of Advanced Materials Research
Vol. 6, No. 2, 2020, pp. 29-36
http://www.aiscience.org/journal/ijamr
ISSN: 2381-6805 (Print); ISSN: 2381-6813 (Online)

Mechanical and Morphological Characterization of Plasticized Low Density Polyethylene (LDPE) Granules Blend with Polyhydroxybutyrate (PHB)

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Abstract

The production and the use of plastics have increased significantly over the last seven decades to meet their ever increasing demand. However, this study examined the mechanical properties of blending polyhydroxybutyrate, PHB and low-density polyethylene LDPE at different proportion with plasticizer. Standard methods were used in the sample preparation and blending procedures. The blends shown a better mechanical properties, elongation at break of the blend were greatly improved, also higher maximum flexural strength of the blends were enhanced. The blends showed higher water uptake than either LDPE or PHB without blending. The composites absorbed water as the days' progresses, no inhibition zone found in the antimicrobial test. The x-ray diffraction (XRD) analysis showed that there is a high similarity in morphology between the composites with lower PHB concentration and the higher ones. Furthermore, the result of degradation test shown that over a short period, the composite could probably degrade completely. *Bacillus substilis* isolated for the study revealed great potential of utilizing the composites, the organisms grew significantly on the composites, the total plate count at the initial stage increased rapidly. Composites arising from the blending of LDPE with PHB could exhibit a better performance than LDPE only in food packaging and other biomedical applications.

Keywords

LDPE, PHB, XRD, Mechanical Properties

Received: July 8, 2020 / Accepted: July 25, 2020 / Published online: August 26, 2020

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1. Introduction

For about a decade ago, production of plastics has been majorly from petroleum which are not biodegradable easily [1]. The environmental persistence of plastic and its incineration which produces toxic gases has led to more research on biodegradable polymers especially from renewable resources especially from agricultural and biomass feedstock which have shown high efficiency to replace feedstock gotten from petroleum resources [2, 3]. These biodegradable polymers are characterized as materials that are degraded by the action of microorganism and the major end-products are carbon dioxide, water and biomass [4]. Among the petroleum based polymer is the Low-Density Polyethylene which is sometimes recycled. It is a very healthy plastic that tends to be both durable and flexible. Items such as cling-film, sandwich bags, squeezable bottles, and plastic grocery bags are made from LDPE. LDPE are tough and flexible, waxy surface, soft – scratches easily and good moisture barrier properties [5]. Poly (3-hydroxybutyrate) (PHB) is a typical natural biodegradable thermoplastic polyester polymer produced by bacterial fermentation and degrades in the environment in few weeks [6]. Its application limitation includes brittleness, inherent

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rigidity, high production cost, and low melt stability but despite all these limitations, PHB have comparable thermal and mechanical properties to those of some conventional polymers and this has generated much interest in exploring their physical and processing properties for potential applications [7, 8].

Blending of polymers is a process of combining different types of existing polymers aiming to achieve intermediate or better properties while preserving the major characteristics of the two pure components and also improve their application [9, 10]. Some blending has been done by some researchers such as Miroslava et al., 2015 [4] on polymer clay nanocomposites that improved its flexibility, compatibility and mechanical properties. Also, significant advancement in morphology and mechanical properties were observed after the additional modified clay was incorporated into the PHB/PCL blend. Mohamed et al. (2012) [11]. Investigated on the thermal and morphology of blended PLA-PHB and the result showed that elongation at break of the blend were greatly improved. Despite great achievements by previous researchers, the persistence of plastics in the environment kept increasing on daily basis due to inability to decompose when disposed and making the environment unsafe for aquatic life, animals and man. Therefore, the focus is to study the mechanical property and morphology of a virgin LDPE blend with PHB for improved properties.

2. Materials and Method

2.1. Materials

Polyhydroxybutyrate (PHB) granules 5mm, 550kg/mol was purchased from Good fellow Cambridge limited (Huntingdom, England), low density polyethylene granules (PE) and plasticizer (glycerol) were purchased from Sigma-Aldrich UK**.**

2.2. Preparation of Blends and Blending Procedures

Polyethylene granules were weighed separately and the percentage of PHB that was needed was worked out together with plasticizer as described by Marisa et al., 2015 [12]. Table 1 shows the composition formulation of the blend. Electronic weighing balance was used to weigh all the materials as stated below.

Solvent casting method was employed in the blending procedure. Exactly 100mL of chloroform was added to the weighed granules of polyethylene and PHB as stated in the table below, temperature was kept at 180°C for 20 min after complete melt of the granules the chamber was opened and condenser gently released and plasticizer was added the mixture was carefully stirred, then poured into stainless plate lubricated with petroleum jelly for easy remover. This procedure was carried out for all the blends formulated. The resulting blends were sun-dried to remove organic solvent left in the blend [13].

Table 1. Composition of LDPE and PHB blends.

Sample (%) PHB	LDPE (g)	plasticizer glycerol (g)
5	17	2
10	16	2
15	15	2
20	14	2
25	13	2

2.3. Mechanical Properties of the Blend

The analysis was done with universal tensile testing machine (Instron-series 5369) Load cell Capacity-50KN. The blended biopolymer was carefully cut without introducing any crack traces. Geometry (Tensile) Gauge length-45mm, Width-10mm, Thickness 5mm. Flexural test, length 80mm, width 25mm, thickness 8mm. Elongation at break, maximum tensile strength, tensile strain and young modulus also maximum flexural strength, flexural strain, young modulus was done simultaneously.

2.4. X-ray Diffraction

The composite blends were cut into smaller pieces and oven dried at 80°C for 5 hrs, samples V/PHB 5, V/PHB 15, V/PHB 25 were grated manually with hand grater and sent for XRD analysis at Centre of Excellence in Nanotechnology and Advanced Materials, National Agency for Science and Engineering Infrastructure (NASENI), Akure, Nigeria. X-ray diffraction (XRD) analyses were performed with a Philips 1820 diffractometer operated at 45 kV and, 40 mA using Cu-Ka radiation with a graphite diffracted beam monochromator. Data were acquired in a 5Φ scale from 10^o to 45^o

2.5. Water Uptake

The samples were cut in a square 1.5 inches, 8mm thickness. Exactly 2g of each test samples were weighed on an electric balance capable of reading 0.0001g. 250mL of clean water were measured and poured into plastic cup and the samples were emerged into the water and kept at 35°C. The samples were removed at intervals of 24h for seven days and weighed.

$$Increase\ in\ weight\% = \frac{Wet\ weight - Conditioned\ Weight}{Conditioned\ Weight}\ X\ 100 \quad (1)$$

2.6. Antimicrobial Test

For the microbial inhibition test, agar disk diffusion method was employed Madalina, *et al.,* 2015 [14]. *Bacillus cereus, Klelsiella Oxytoca, Staphlococcus, Escherichia Coli and*

Salmonella typhimurium were used for the study. The isolates were inoculated in peptone broth on the Muller Hinton agar plate using spread plate techniques. The test plastics (blend) were cut into small sizes (1cm * 1cm) and placed on inoculated plates to check their antimicrobial properties. The plates were incubated at 37°C for 24 h and the inhibition diameters measured afterward.

2.7. Soil Degradation

The loamy soil used for this assay consists of 20% sand, and 80% loam. Notably, 2g of the test composite were used, while 1000g (1kg) soil per pot were weighed the experiment were carried out in three testing periods (30 days, 60days and 90days). Sample were buried 2cm beneath the surface under laboratory conditions during the test period, the soil was regularly irrigated with clean water to maintain a stable humidity. After a predetermined degradation time, the sample were carefully removed from the soil to avoid damage, cleaned with water and dried. The dried samples (films) were weighed to calculate the weight loss as described by Rychter et al., 2010 [15].

$$\%\, Massloss = \frac{Mi-Mf}{Mi} X\ 100 \qquad (2)$$

2.8. Degradation in Slurry Condition

A pure strain of *Bacillus substilis* on agar slant was obtained from the research laboratory in the Department of Microbiology, Federal University of Technology, Akure. The collected *B. Substilis* was purified by streaking severally on nutrient agar. Following purification, broth cultures of the test organisms were prepared aseptically in nutrient broth and incubated for 24 hrs at 37°C, after 24 hrs of growth, the culture media was used for biodegradation study. 2 g of the films (V/PHB5, V/PHB10, V/PHB15, V/PHB20, and V/PHB 25) were aseptically weighed using a weighing balance and kept for further use. After 24 hrs growth, 10 mL of a known concentration of *B. substilis* was introduced into sterile containers containing 100 mL sterile nutrient broth each and the known weighed test samples to be degraded was added. The set-up was incubated at 50 rpm for 21 days at room temperature. The direct viable bacterial count was carried out after every 24h for 7 days to monitor the concentration of *B. substilis*. Bacterial colony counts were recorded as colony forming unit per milliliter (cfu/mL). At the end of 21 days, the test samples were brought out and weighed and then recorded. Likewise, the direct viable bacterial count was also carried out after the 21 days of the experimental set-up and bacterial colony counts were recorded as colony forming unit per milliliter (cfu/mL)

3. Result and Discussion

3.1. Mechanical Properties

3.1.1. Tensile Test

The tensile test is shown in Table 2. Tensile strength value of Virgin PHB was 4.5209 MPa, which was in the range of commercial PHB reported by Ch'ng and Sudesh, 2013 [16, 12] and 5.9860 MPa for the Tensile strength of Virgin LDPE. In contrast, Gonzalez et al 2014 [17] studied blends of linear low density polyethylene (LLDPE) and ethene-propene-1-butene copolymer with different compositions and found tensile strength of 23 MPa for LDPE, higher than the values of the LDPE evaluated in this work, of 5.9860 MPa.

Tensile strength of PHB/ LDPE blends ranges from 6.1933 to 7.874 MPa with best at 7.8741 MPa, whereas elongation at break varied from 12.83 to 65.57% with 65.57%. It was evident from results that blending had improved tensile propertied due to intermolecular bonding and increase in amorphous content [18]. The incorporation of PHB into Virgin LDPE did not follow a consistent pattern or manner to which the properties are been increased but the maximum tensile strength and young modulus was found in the VIG/15PHB (15 g VIRGIN LDPE, 3 g PHB, and 2 g plasticizer). This suggests that maximum tensile strength impact is not a factor of the quantity of PHB added rather it is the blending ratio that influences the tensile strength.

Table 2. Tensile Strength of LDPE/PHB blends.

Samples	Elongation at Break	Maximum Tensile strength (mPA)	Tensile strain (Standard) (mm)	Young Modulus (E)
PHB	29.24 ± 0.523	4.5209 ± 0.002	0.2402 ± 0.001	10.453 ± 0.028
LDPE	36.24 ± 0.684	5.98650 ± 0.002	0.3425 ± 0.001	17.47883 ± 0.027
V/PHB5	35.33 ± 0.425	6.19331 ± 0.003	0.35333 ± 0.001	17.5284 ± 0.025
V/PHB10	23.04 ± 0.322	8.80691 ± 0.028	0.23043 ± 0.001	38.21946 ± 0.027
V/PHB15	12.83 ± 0.021	9.73107 ± 0.027	0.12833 ± 0.001	75.82849 ± 1.230
V/PHB20	55.83 ± 0.987	9.14527 ± 0.029	0.55833 ± 0.002	16.37969 ± 0.034
V/PHB25	65.57 ± 1.002	7.87416 ± 0.025	0.65570 ± 0.002	12.00878 ± 0.042

3.1.2. Flexural Test

The flexural test results were presented in Table 3 for virgin polyethylene blend with PHB. The result showed that for the virgin polyethylene blend, the maximum flexural strength ranges from 3.75345 mPA to 17.08062 mPA, flexural strain

ranges from 2.11245 mm to 10.02675 mm while the young modulus range from 0.56601 to 8.08569. The result of the polyethylene-PHB blend shows that all form of the blends had higher maximum flexural strength than their respective parent polyethylene. It suggests from results that blending had improved maximum flexural property due to intermolecular bonding and increase in amorphous content [18]. It was observed that in the polyethylene blend, an increase in the maximum flexural strength and young modulus resulted into corresponding reduction in the flexural strain but was not constant indicating that improvement in the flexural strength improvement can be achieved by appropriate combination of the mixture of PHB and polyethylene.

Table 3. Flexural Test of LDPE/PHB blend.

Samples	Maximum Flexural strength (mPA)	Flexural strain (Standard) (mm)	Young Modulus (E)
VIGLDPE	3.75345 ± 0.023	6.5526 ± 0.024	0.572818 ± 0.005
V/PHB5	3.85901 ± 0.027	6.80909 ± 0.054	0.566744 ± 0.002
V/PHB10	17.08062 ± 0.523	2.11245 ± 0.007	8.085692 ± 0.523
V/PHB15	12.21704 ± 0.022	4.45418 ± 0.001	2.742826 ± 0.023
V/PHB20	9.95369 ± 0.523	10.02675 ± 0.724	0.992713 ± 0.003
V/PHB25	4.67434 ± 0.033	5.02367 ± 0.001	0.930463 ± 0.003

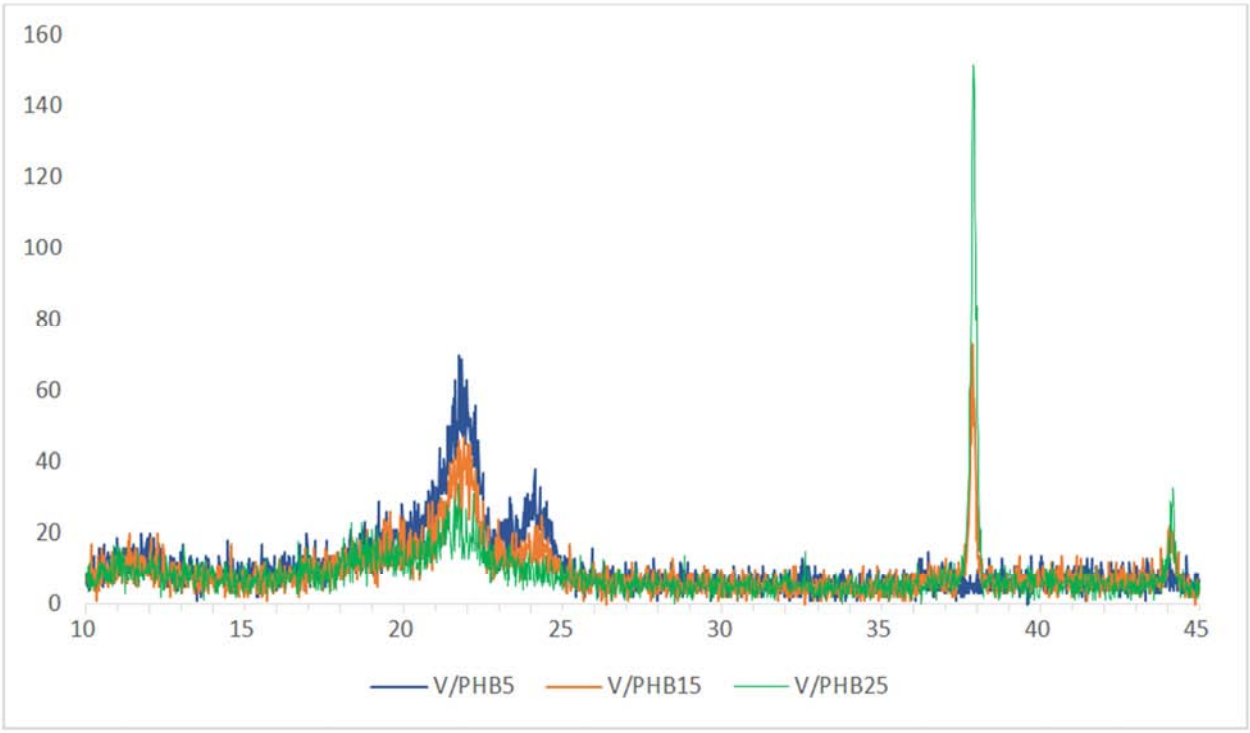

Figure 1. X-Ray diffraction patterns of PHB blends with LDPE films.

3.2. XRD Analysis

Figure 1 shows the XRD result of 5%, 15% and 25% sample. Despite the different composition of the composites, similar diffraction peaks were observed at nearly the same position indicating that these composites have typical crystal structures. PHB is a highly ordered polymer and is known to crystallize in an orthorhombic cell [19,20]. In general, the patterns of LDPE and PHB blends are very similar to that of standard neat PLA and neat PHB [11]. The development of crystallinity, improved with the plasticizer content in the films two diffraction peaks at 20° and 23° were observed. The first one corresponds to the characteristic peak of PHB crystallinity, while the other peak overlapped at 24° was attributed to the presence of LDPE. The intensity of these peaks increased at 38° and it became more defined. Furthermore, these results suggest that the presence of plasticizer improved the interaction between LDPE and PHB and consequently the development of crystallinity in the blends structure [21].

3.3. Water Uptake

The result of the water uptake is presented in Figure 2. The result shows that all blends have higher water uptake than the Virgin LDPE and PHB only. In the Virgin LDPE blends, the highest water uptake was found in the VPE+PHB20 while the lowest with LDPE+PHB5. This suggests that the concentration of PHB in the blend has significant effect on the water uptake of the blend. As the time progresses, the composites absorbed water while some of the composites particles were seen in the water. This presence of composites particles in the aqueous medium might be attributed to the PHB in the blends which could also suggested possible degradation if by any means it gets into water environment [22].

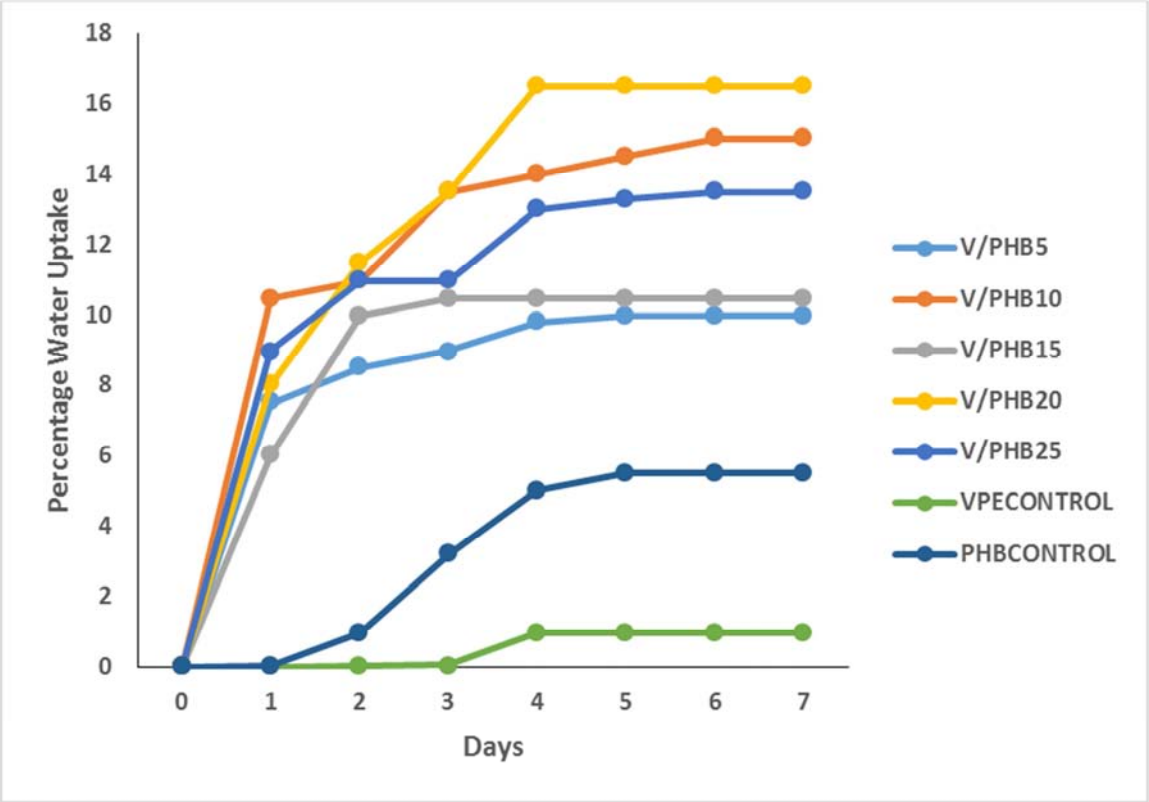

Figure 2. Water uptake of PHB blend with LDPE.

3.4. Antimicrobial Test

The antimicrobial activity of the test films shows no inhibition zone around the polymers films, the isolates grew significantly, no clear zones on the plastic-infused media, implying that the microbes could probably possess plastic degrading enzyme machinery. It measured 0.00 mm for all blends.

Figure 3. Antimicrobial Test of the blend.

3.5. Soil Degradation of Polyhydroxybutyrate Blend with Virgin Polyethylene

The degradation test after 90 days showed that the LDPE control is an inert polymer with strong resistance to microbial breakdown as such it does not degrade. The degradation results showed increases with increase in concentration of PHB added compare to the PHB control which increase significantly. Weight loss increase as the number of day's increases from 30 days to 90 days which implies that PHB blends with LDPE will be biodegradable plastics provided right proportion is used [23].

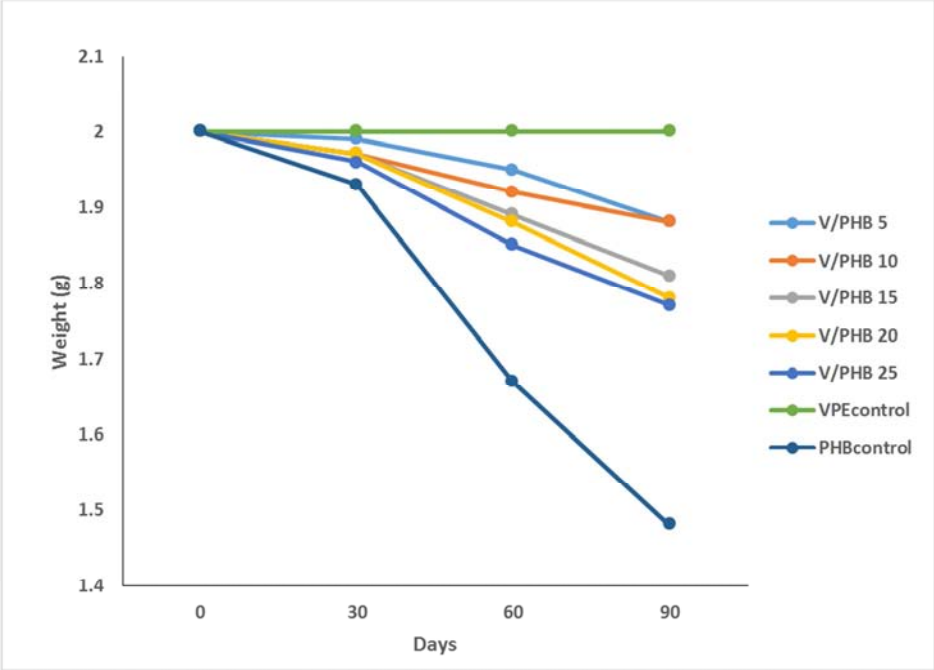

Figure 4. Soil degradation of PHB blends with LDPE.

3.6. Slurry Degradation

Screening the isolates (*bacillus substilis*) for bio plastic degradation revealed that the isolates were capable of utilizing plastics as a carbon source. According to the results shown, there was increase in the total plate count at the initial stages due to increase in the microorganism population. However, the population of the microbes reduced as the test films diminished, which might be as a result of insufficient nutrients to feed on, as the microbes compete for nutrients, space and survival. The smaller the films, the smaller the population of microbes present in the test films. The final dry weight showed that there was reduction in weight of the blends which indicate that the microorganism has actually degraded the polymer. Furthermore, there was reduction in the weight of the polymer containing the highest concentration of PHB. This study suggests that the incorporation of PHB into synthetic plastics could make the resulting composite material be more biodegradable, thus making it a bio plastic of choice for greener environment.

Table 4. Microbial count and degradation in Slurry.

TEST SAMPLES	WBB grams	DAY 1 TPC (cfu/mL)	DAY 2 TPC (cfu/mL)	DAY 3 TPC (cfu/mL)	DAY 4 TPC (cfu/mL)	DAY 5 TPC (cfu/mL)	DAY6 TPC (cfu/mL)	DAY 7 TPC (cfu/mL)	DAY 21 TPC (cfu/mL)	wWAB grams	dWABgrams
PHB5	2.00	$2.06x10^5$	$2.77x10^5$	$4.45x10^7$	$4.00x10^7$	$1.80x10^7$	$1.08x10^7$	$0.77x10^7$	$1.05x10^2$	1.26	0.74
PHB10	2.00	$1.86x10^5$	$2.66x10^5$	$3.93x10^7$	$3.90x10^7$	$1.80x10^7$	$1.10x10^7$	$0.80x10^7$	$1.02x10^2$	1.29	0.71
PHB15	2.00	$2.01x10^5$	$2.81x10^5$	$4.40x10^7$	$4.10x10^7$	$2.10x10^7$	$1.28x10^7$	$0.92x10^7$	$1.04x10^2$	1.16	0.84
PHB20	2.00	$1.99x10^5$	$2.66x10^5$	$4.28x10^7$	$2.70x10^7$	$2.04x10^7$	$1.22x10^7$	$0.80x10^7$	$1.09x10^2$	1.02	0.89
PHB25	2.00	$1.80x10^5$	$2.70x10^5$	$4.45x10^7$	$2.98x10^7$	$1.01x10^7$	$1.08x10^7$	$0.96x10^7$	$0.08x10^2$	1.21	0.76

Keys:
WBB = Weight before Biodegradation
*w*WAB = wet Weight After Biodegradation
*d*WAB = dry Weight After Biodegradation
TPC = Total Plate Count
Cfu/ml = Colony forming unit per milliliter

4. Conclusion

The PHB loadings at different proportions were blended with LDPE. The result of the mechanical test shows that both for the tensile strength and flexural strength were increased with PHB loadings. Also it was noted that the strength of the LDPE used for the blend improve the functional properties of the composites. In addition, the tensile strength was not based on the maximum concentration of PHB added but the right formulation with the LDPE. The blends also showed better water uptake than the Virgin PHB and LDPE. Practical implication of this study suggest its use in the production of green plastics that are environmentally sustainable.

Declarations

Funding

By Babaniyi Babafemi Raphael.

Conflict of Interest

Not Applicable.

Availability of Data and Material

Not Applicable.

Code Availability

Not Applicable.

Authors Contribution

This study was collectively done by the authors. Author BBR conceived the idea, carried out the experimental work and interpreted the data. Author AOA supervised and edited the manuscript. All authors read and approved the manuscript.

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