Characterization study of cellulose (CEL) + plasticizer PEG added LDPE bio-composite.

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Abstract

The development of society and increasing consumption of plastics in the world has been a subject of great concern and special attention by the scientific community. The aim of this research paper to synthesize and characterize Cellulose + PEG added LDPE bio composite thin film buried in natural compostable soil environment for the period up to 180 days. The Cellulose (CEL) added 7 wt.% PEG/LDPE bio composite thin films having 10 wt.% of CEL were characterized by weight loss and weight loss percent, UTS and PEB measurements, SEM and XRD before and after degradation in order to study biodegradation of CEL/PEG/LDPE thin film samples under natural environment. The LDPE + 7% PEG + 10% CEL bio composite thin films after soil burial of 90, 120, 180 days are found to be degraded and makes an eco- friendly and degradable material in natural environment.

Keywords: Biodegradation, LDPE + PEG + CEL, Soil burial, XRD, SEM, UTS, PEB

Accepted on February 22, 2021

Introduction

Plastics have been an integral part of our lives for several decades in the form of convenient and useful commodity items. Plastic products are usually inert, chemically stable, resistant to corrosion, water-proof, durable and light-weight, ironically, the same features which make them ideal as a raw material for such a wide range of products are also responsible for the environmental problems cited by many environmentalists. Plastics are synthetic, semi synthetic and natural. Environmental pollution by synthetic polymers, such as waste plastics has been recognized as major problem. In view of this, energetic, chemical and biological polymer degrading techniques have been studied extensively during last three decades [1]. Usage of certain microorganisms and enzymes to degrade polymers are classified as the biodegradation method of polymer. The biodegradability of plastics is depends on the chemical structure of the material and on the constitution of the final product, not just on the raw materials used for this production. The degradation of conventional plastics such as polyethylene, polypropylene, polystyrene, poly (vinyl chloride) and poly (ethylene terephthalate) by the action of natural herbs polymers such as starch [2], cellulose, lignin, chitin [3] is a significant way to accelerate polymer biodegradation. Microorganism breaks the polymeric chain and consumes materials through aerobic and anaerobic process. Research is now going towards in replacing some or all kind of synthetic polymers such as PE, to become biodegradable materials due to their pollution in environment.

LDPE is one of the most widely used packaging plastic, is the worst offender, being highly resistance to degradation [4]. Polyethylene glycol (PEG) is non-hazardous and biocompatible additive to support degradation. During degradation, the PEG molecules are reduced by one glycol unit at a time after each oxidation cycle and Cellulose (CEL) is organic material likely consumed by microorganisms which enhances the rate of degradation in compostable soil environment. Thus, the aim of the present investigation to study the biodegradation of CEL + PEG added LDPE composite thin film in compostable soil (Soil and compost is in 1:1 proportion) burial natural environment.

Materials and Methods

Commercial grade LDPE with density 0.924 g/cm^3 at 23 ^{0}C, melt flow index 4 gm/10 min; melting range 105-109^{0}C was supplied from Nutan Gujarat Industrial Estate, Vadodara (India). Xylene as solvents of AR Grade and PEG with average molecular weights 3500-4000, freezing point 53-50^{0}C and having pH in between 4.5 to 7.5 is used as provided without further purification. Cellulose powder with pH in between 5.0 to 7.0 obtained from Lobe Chemicals, Mumbai. The compostable soil (1:1 proportion) has pH 8.43 before and pH 8.34 after 180 days.

Sample preparation

The Cellulose+PEG added LDPE composite thin films were synthesized by solution evaporation technique [5]. Concentration of Cellulose (10 wt.%) and PEG (7 wt.%) in LDPE and the sample is coded as LP7P1^{0}C0 before degradation and the same sample after 90 (LP7P1^{0}C9), 120 (LP7P1^{0}C12) and 180 (LP7P1^{0}C18) days of degradation respectively. The thickness of the film is in range of 15-30 μm. A German make microscope supplied by Paul Waechter, Model No. 641640 was used for thickness measurement [6].

Soil burial technique

A soil burial technique is carried out in natural environment to degrade the sample. For this, a thin rectangular sheet of sample (about 2.5 cm × 12 cm) of 15-30 µm thickness is buried at the depth of 5cm in compostable soil having pH 8.43 before and pH 8.34 after 180 days. The compostable soil having pH 8.43 (with N=588.0, P=27.06, K=249.09) before burial of samples and pH 8.34 (with N=280.0, P=30.38, K=192.06) after 180 days. The moisture content was maintained by adding water in soil after regular interval. The samples were removed from soil after 90,120 and 180 days and washed with distilled water and then acetone bath and dried at room temperature. The samples were characterised by weight loss and weight loss percentage, UTS and PEB measurements, SEM and XRD.

Mechanical testing

Mechanical properties of samples LP7P1^{0}C0, LP7P1^{0}C9, LP7P1^{0}C12 and LP7P1^{0}C18 (about 2.5 cm × 12 cm) having 15-30 µm thickness were carried out using an ASTM D 882 Universal Testing Machine(UTM) , Nova Surface-Care Centre Pvt. Ltd. Mumbai. Ultimate Tensile Strength (UTS) and Percentage Elongation at Break (PEB) before and after degradation were measured. Tensile strength represents the capacity to take load by film and percentage elongation at break is its ability to stretch. Mechanical behaviour of polymer affected by many factors like molecular weight, molecular orientation, branching, crystallinity and density, processing conditions and processing techniques and also the thickness of film.

Result and Discussion

Weight loss and weight loss percent study

Weight loss of all CEL(10 wt.%) + PEG(7 wt.%) added LDPE thin film samples determined from the weight measurement before and after degradation for particular time period and carried out on K-Roy Monopan Balance having accuracy + 0.1 µgm and are enclosed in Table 1.

Table 1. *Weight loss and weight loss percent of LP7P10C0, LP7P10C9, LP7P10C12 and LP7P10C18 samples.*

Sample Code (days)	Weight (mg)	Weight Loss %
LP7P10C0 (0 day)	1.84	0
LP7P10C9 (90 days)	1.57	14.67
LP7P10C12 (120 days)	1.59	13.59
LP7P10C18 (180 days)	1.55	15.76

Burial in compostable soil. But after 90, 120 and 180 days of compostable soil burial, the weight of thin film sample decreases and it becomes 1.570 mg for (LP7P1^{0}C9), 1.590 mg for (LP7P1^{0}C12) and 1.550 mg for (LP7P1^{0}C18) samples. In terms of weight loss percent it becomes 14.67%, 13.59% and 15.76% for 90, 120 and 180 days of degradation respectively. Thus weight loss percent was found to increase after 90 and 180 days but slight weight loss after 120 days. The growth of microorganisms within the polymer leads to an increase in weight where as a loss of polymer integrity leads to a weight loss. The weight loss is proportional to surface and biodegradation initiated at the surface of the polymer. Many researchers adopted this method for the evaluation of degradation [7].

Tensile strength and percentage of elongation break

Measurement of mechanical properties such as Ultimate Tensile Strength (UTS) and Percentage Elongation Break (PEB) of samples LP7P1^{0}C0, LP71^{0}CP9, LP7P1^{0}C12 and LP7P1^{0}C18 carried out using an ASTM D 882 Universal Testing Machine (UTM), Nova Surface-Care Centre Pvt. Ltd. Mumbai.

Ultimate tensile strength of LP7P1^{0}C0 (0 days) sample found to be 2.31 MPa and percentage elongation at break value found to be 3.48% before degradation. But when thin composite sample of 10 wt.% CEL added 7 wt.% PEG in LDPE enter into compostable soil environment, its UTS value gradually decreases with increases weathering duration and reduced to 40.26% after 180 days of degradation. PEB value of 10 wt.% CEL added 7 wt.% PEG in LDPE composite sample found to be increase after 90 days of degradation and then decreases after 120 and 180 days of degradation and reduced to 74.42% of before weathering.

Elongation is a more useful measure of oxidative degradation than tensile strength. Tensile strength may not be markedly affected by sample embrittlement, whereas, elongation is much more sensitive and can provide evidence of polymer degradation. Chain scission and cross linking reactions in polymer are the direct cause of decrement in tensile strength [8].

Scanning electron microscopy (SEM) analysis

SEM of all thin film samples were recorded on scanning electron microscope (SEM, ZEISS EVO18), R.T. M. Nagpur University, Nagpur (India). Photograph of LP7P1^{0}C0 (0 day) show plane transparent heterogeneous structure with traces of cellulose (CEL) interspersed in thermoplastic matrix as filler (Figure 1).

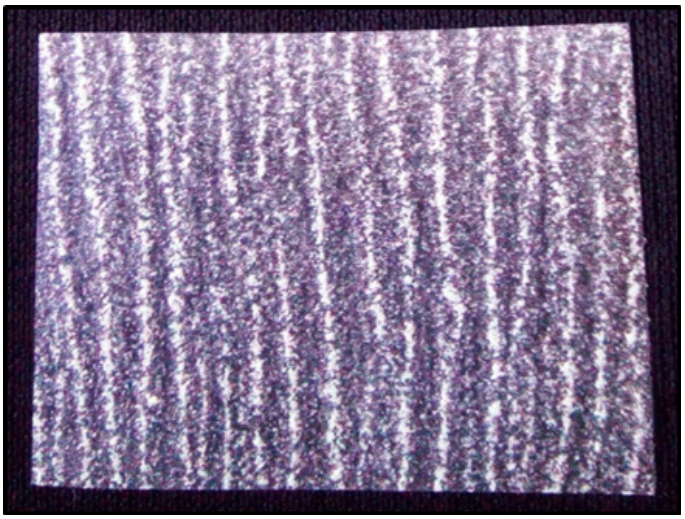

Figure 1*. Photograph of LP7P1^{0}C0*

In SEM of LP7P1^{0}C9 small circles or pores of irregular shape and size are observed on the surface of thin film after 90 days of compostable soil degradation. These pores created by micro-organisms present in compostablet soil. After 180 days of compostable soil degradation, SEM of LP7P1^{0}C18 bigger pores of irregular shape and sizes as a result of biological degradation. These results show that, CEL which is loosely bounded with PEG/LDPE matrix was first attacked by the microorganisms

such as fungi and bacteria due to moisture absorption and get hydrolysed.

Cellulose (CEL) is odourless, no taste, hydrophilic, insoluble organic in water compound, a polysaccharide consisting of a linear chain of several hundred to many thousands of linked D-glucose units. SEM micrographs of LDPE-cellulose revealed that, addition of plasticizer (PEG) to composites enhances their dispersion in continuous LDPE phase and facilitates direct contact between cellulose fiber and LDPE matrix. Tensile strength of LDPE-Cellulose added PEG composites decreases as cellulose content increases [9]. The above result supports the mechanical properties result of CEL/PEG/LDPE sample.

X-ray Diffraction (XRD)

X-ray diffraction (XRD) data of all thin film samples were recorded on Bruker AXS D8 Advance, STIC Cochin University, Cochin, Kerala (India).

The XRD patterns for 10 wt.% CEL/7 wt.% PEG/LDPE ($LP7P1^0C0$, $LP7P1^0C9$, $LP7P1^0C12$ and $LP7P1^0C18$ before and after degradation respectively). From XRD pattern of $LP7P1^0C0$ (0 day) sample shows first prominent peaks at $2\theta = 22.030$ and second small peak at $2\theta = 24.280$ corresponding to d-spacing of 4.03 A0 and 3.66 A0 respectively. But when 10 wt.% CEL/PEG/LDPE samples entered in compostable soil environment for 90 ($LP7P1^0C9$) , 120 ($LP7P1^0C12$) and 180 ($LP7P1^0C18$) days, after degradation it is observed that XRD peak shifts to higher 2θ values for all degraded samples and corresponding d-spacing values decreases with increasing days of degradation. After weathering of 10 wt.% CEL/7 wt.% PEG/LDPE degraded samples it is observed that, intensity of XRD peak slight increases after 90 and 180 days of degradation and decreases after 120 days of degradation. Decrement in d-spacing and intensity of XRD peak with weathering duration in compostable soil decreases crystallinity and support biodegradation of CEL/PEG/LDPE samples. Our findings are in good agreement with many researchers [10].

Weight loss and weight loss percent study found that weight of 10 wt.% CEL + 7 wt.% PEG added LDPE samples decreases with increasing days of weathering duration in compostable soil and weight loss percent was found to increase after 90 and 180 days but slight weight loss after 120 days. Here the weight loss is proportional to surface and biodegradation initiated at the surface of the polymer. Mechanical properties result shows that ultimate tensile strength (UTS) value gradually decreases with increases weathering duration and reduced to 40.26% after 180 days of degradation. PEB value of 10 wt.% CEL added 7 wt.% PEG in LDPE composite sample found to be increase after 90 days of degradation and then decreases after 120 and 180 days of degradation and reduced to 74.42% of before weathering sample. Chain scission and cross linking reaction in polymer are the direct cause of decrement in tensile strength. The percentage of elongation break decreases gradually with increasing degradation duration. SEM results indicate that shape of circles or pores of irregular shape and size are observed on the surface of thin film increases with burial duration. These pores created by micro-organisms present in compostable soil. Hence after 180 days of compostable soil degradation, SEM of $LP7P1^0C18$ shows bigger pores of irregular shape and sizes as a result of biological degradation. These results indicate that, CEL which is loosely bounded with PEG/LDPE matrix was first attacked by the microorganisms such as fungi and bacteria due to moisture absorption and get hydrolysed. This result supports mechanical property results. XRD results shows that decrease in crystallinity of 10 wt.% CEL + 7 wt.% PEG/LDPE thin film samples with increase in weathering days in compostable soil indicates increase in amorphous phase of thin film sample which suggest that degradation of CEL/PEG/LDPE samples increases with weathering duration.

Conclusion

The Weight loss, UTS, PEB, SEM and XRD study of LDPE+7%PEG +10% CEL degraded (for 0, 90, 120, 180 days) sample revealed that addition of Cellulose (CEL) along with PEG affect nature and structure of LDPE and enhances biodegradation with days of degradation.

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