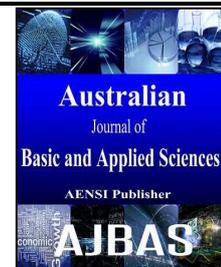




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Preparation and properties of Biodegradable Blend from LDPE/PVA Mixture

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ABSTRACT

Polyethylene (PE) and polypropene (PP) are some of the most dominant packaging materials and responsible by part of the problems in the disposal of one-trip packaging. They are high hydrophobic level, water repellence and high molecular weight and their lack of functional groups recognisable by microbial enzymatic systems. Being hydrophobic hydrocarbon polymers, polyolefins are resistant to hydrolysis and for this reason they cannot hydrobiodegrade. Polyolefins, as commercial products, are also resistant to oxidation and biodegradation due to the presence of anti-oxidants and stabilizers. Biodegradable materials derived from renewable resources have been the center of public interest for environmental protection and sustainable development. Biodegradable polymers are, however, suitable in many commodities and medical applications such as packaging, surgical implants, controlled release, and drug delivery systems although their use is still limited because of their high cost or low performance. The current research aims to prepare polymer blend have the ability to partial decomposition when burial the waste in soil, which can used for some applications such as packing and agricultural to solve solid waste problems. The samples are prepared by using twin screw extruder. LDPE and PVA has been mixing with different weight proportions, and studied their properties in order to determine its compliance with the required specifications to be able to be used biodegradable polymers. To improve the viability of decomposition PEG has been added to the resulting blend. Several tests were applied to identify those properties such as tensile, hardness, density and creep test. FTIR, digital microscope and SEM test achieved in order to determine the miscibility and blend morphology before and after degradation. The results show that, the mechanical properties of the blend decrease with increase PVA content. Based on a soil burial test the biodegradability of the blends show clear trend of increase degradation with increasing PVA content.

INTRODUCTION

An important alternative to minimize the polymer waste is the introduction of biodegradable polymers, which can be degraded through the action of naturally occurring microorganisms (Samarasekara, A. and E. Jayasuriya, 2013). There is a considerable interest in replacing some or all of the synthetic plastics by natural or biodegradable materials in many applications, such as, food industry and agricultural which produce a lot of quantity of waste (Behjat Tajeddin, 2009).

The recent incorporation of biological waste treatment (i.e., composting and biogasification) in an integrated approach to solid waste management has resulted in a growing commercial interest in the development of biodegradable materials for consumer products (White, P., *et al.*, 1994; Van Der Zee M., 1997). There are a number of materials known as biodegradable plastics (i.e., starch-based materials, cellulose-derived polymers, bacterial polyesters and a range of synthetic polymers). The main problem associated with designing

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biodegradable polymers is the optimization of their chemical, physical and/or mechanical properties, as well as their biodegradability (Van Der Zee M., 1997; Fritz, H.G., *et al.*, 1994).

In section is present in enough quantities and if that if is the biodegradable disposal environment degrade it, the plastic microorganisms in the waste inert component should lose its integrity, or film containing the residual best- renewable resources able to fall to pieces and fade away. The create biopolymer and biodegradable plastics are starch and cellulose (Behjat Tajeddin, 2009).

Nowadays, the blending of biodegradable polymers with inert polymers has been accepted as a possible application in the waste disposal of plastics. In principal, the way of thinking behind this method is that if the biodegradable section is present in enough quantities and if microorganisms in the waste disposal environment degrade it, the plastic or film containing the residual inert component should lose its integrity, fall to pieces and fade away. Natural biopolymers including starch, cellulose, and chitosan were tested, alone or combined with synthetic polymers, to explore the possibility of forming a fully or partially biodegradable film (Yeon-Hum Yun, Soon-Do Yoon, 2010; N. A. Azahari, N. Othman and H. Ismail, 2011).

Blending LDPE with biopolymers inserts hydrophilic groups that lower the surface energy. Consumption of the polar hydrophilic biopolymer causes fracture in the polymer chain and, through the fractures, microorganisms can access the carbon links of polyethylene; the result is the polymer biodegradation (Rutkowska, M., *et al.*, 2002).

2. Materials and Sample Preparation:

Low density polyethylene (LDPE) was obtained from Amir Kabir Petrochemical Company as granular materials, Iran. Polyvinyl alcohol (PVA) was obtained from Yonghui chemical Holdings Limited Company, china. Polyethylene glycol (PEG) was obtained from Sinopharm Chemical Reagent Company, Ltd, Chain.

LDPE/PVA blends contain varying ratio according to table1, then PEG added for all sample above also show in same table. The mixing process was carried out in twin screw extruder model (SLJ30A) the mixed materials were fed from a hopper on the screw. It's then conveyed along the barrel where it is heated by conduction from the barrel heaters and shear due to its movement along the screw flight. The depth of the twin screw channel is reduced along the length of the screw so as to compact the material. At the end of the extrusion, the melt passes through a die in the form of sheet. The extruder operation conditions were 35rpm in the beginning increased to 50 rpm of screw speed. and the temperatures used for zones 1 and 2 were (135-150)°C respectively.

Table 1: percent ratio of samples

LDPE%	PVA%	PEG%
100	0	0
90	10	0
80	20	0
70	30	0
60	40	0
85	10	5
75	20	5
65	30	5
55	40	5

3. Apparatus:

Tensile tests is performed on the sample were cut according to the ASTM D 1708-02 specimen dimensions, by machine used for the testing of tensile properties is microcomputer controlled electronic universal testing machine model (WDW-5E), to determined tensile strength and the percentage of elongation .

The creep test is performed on the sample were cut according to the ASTM 2990

The Fourier transform infrared (FTIR) spectra were recorded on a FTIR 8400S-Perkin-Elmer spectrophotometer. The samples were pressed into tablets with potassium permanganate KBr.

Density test is performed using (Matsu Haku, China, HIGH Precision DENSITY TESTER GP-120S with digital accuracy = 0.0001 g/cm³). Which contain water at room temperature and the measure based on Archimedes low.

Digital microscope is performed on the blended samples (model AM4815T Dino-Lite Edge) to show the morphology of the blend.

RESULTS AND DISCUSSION

4.1 Tensile tests results:

Figure 1. Show the effects of polyvinyl alcohol (PVA) and polyethylene glycol (PEG) on the tensile strength for low density polyethylene. The results show that the tensile strength decreases with (PVA) percent increase due to the poor interfacial adhesion between the two polymers with different polarities which results in poor stress-transfer between the matrix and the dispersed phase. It is believed that PVA formed agglomerates due to the strong intermolecular hydrogen bonds between the hydroxyl groups which result in its poor dispersion in LDPE, particularly at higher PVA contents, this agreement with Hanafi Ismail (Hanafi Ismail, *et al.*, 2010).

PVA is highly hydrophilic as it contains hydroxyl groups On its surface whereas LDPE is nonpolar. Therefore, in such a system, the formation of strong interfacial bonds like hydrogen bonds is not feasible this is because at higher PVA contents, PVA-PVA interaction becomes more pronounced than PVA-PE interaction, this agreement with Francis Vidya (Francis, Vidya, 2013).

The effect of addition 5% polyethylene glycol to LDPE/PVA blend show also in figure 1. The results show the tensile strength more decrease with presences (PEG) due to the poor interfacial adhesion between LDPE and PVA The presences PVA and PEG work as plasticizer materials for LDPE which increase chains mobility or sliding one on others, this lead to decreasing in tensile strength.

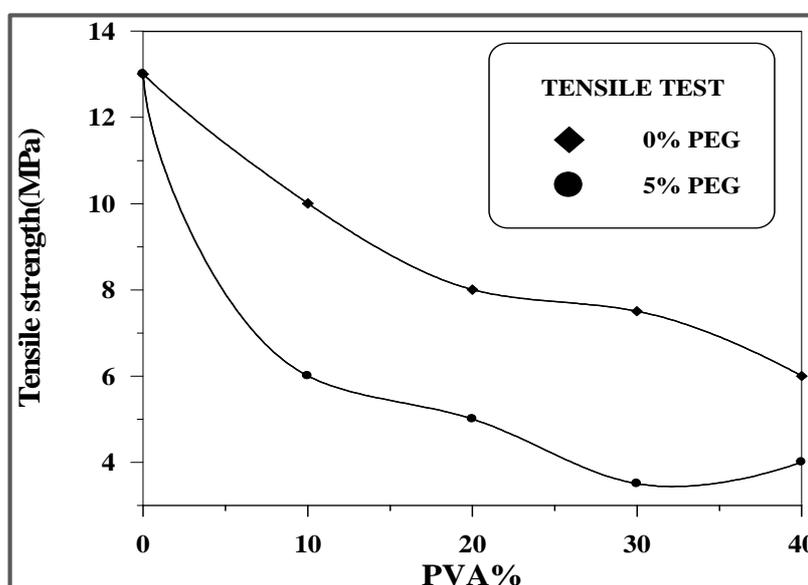


Fig. 1: show effect of PVA percent on the tensile strength of LDPE

The elongation at break (Fig.2) also shows the same trend as that of tensile strength. The drop in elongation of the blends becomes more drastic as the PVA content is increased. Several researchers also report similar trends on mechanical properties of incompatible blends the absence of a compatibilizer resulted in poor adhesion between PVA and LDPE matrix. This leads to easier crack propagation Francis Vidya (Francis, Vidya, 2013).

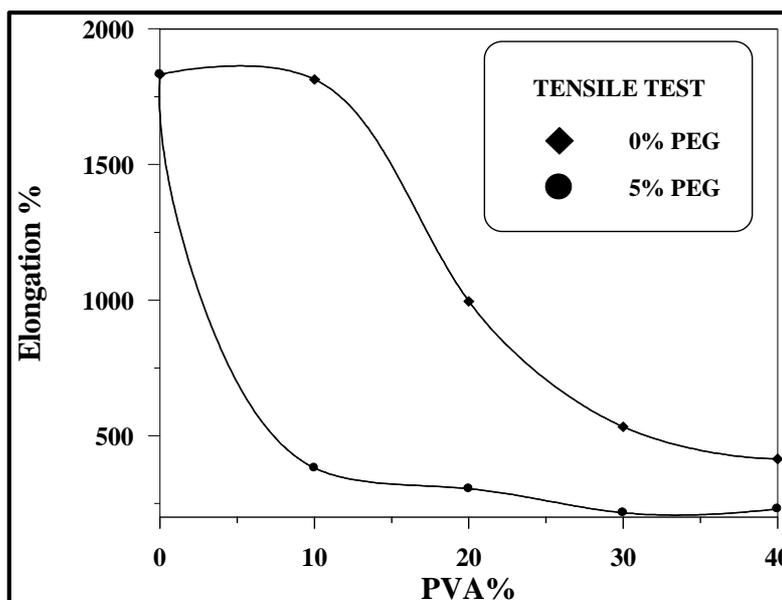


Fig. 2: Effect of PVA percent on the elongation of LDPE

Fig.3 shows an increase in elastic modulus of the blends with increase in PVA content. This results due to the stiffening effect of PVA, which it is stiffer than LDPE. The hydrogen bonds in PVA give it a much higher modulus than semi crystalline polymers such as LDPE which have no hydrogen bonding. Hence the elasticity of the blend falls and the average stiffness increases as PVA tends to agglomerate within the LDPE matrix to form three dimensional reticulate structures Francis Vidya (Francis, Vidya, 2013).

This figure show different behavior at high percent of PVA, this due to increasing agglomerating the PVA between LDPE chains and reduce the chains bonds. The same result with presence (PEG).

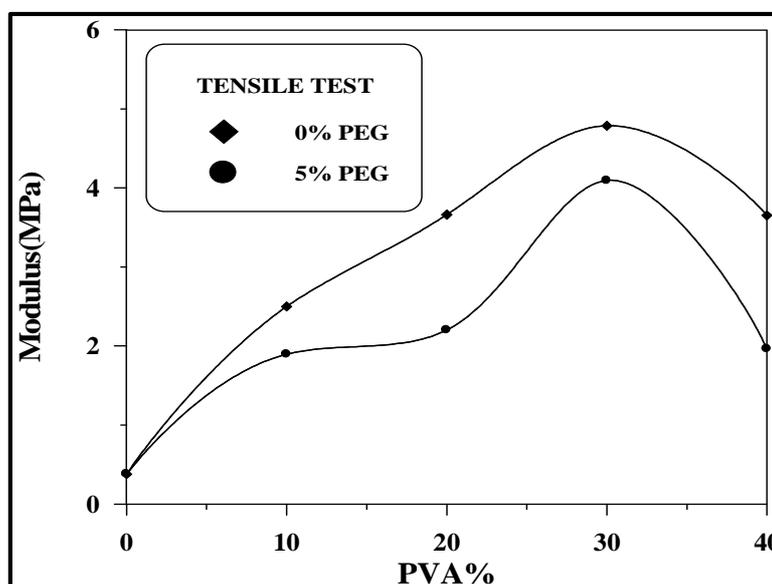


Fig. 3: show effect of PVA percent on the value of modulus of LD

4.2 Creep test result:

Creep test achieved at room temperature with constant load (5 KN) show in Figs.4 and Fig.5 of LDPE/PVA and LDPE/PEG/PVA blends respectively. These figures indicate that the creep rates increase rapidly with the time for all samples, it can also be observed that the creep rates depend on the PVA content, which increases with increasing PVA percent, this due to PVA helps to chains sliding by influence on secondary bonds of LDPE. Changes in behavior, which appeared in this test as a result of the sample subjected to the longest test time, which allows for sliding chains one on others and prevent them from return into previous state.

With an increase PVA, creep rate is reduced due to agglomeration PVA because of the hydrogen bonds that prevention chains from sliding on the other loathed under the influence of creep load.

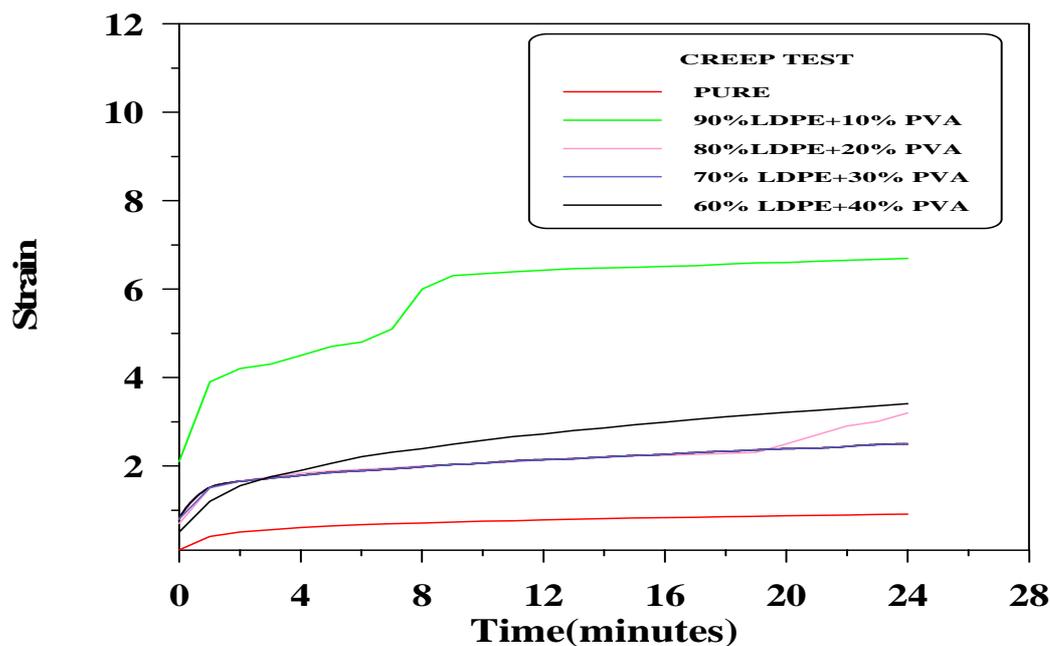


Fig. 4: Show (strain-time) curve of pure LDPE and LDPE with PVA in different percent.

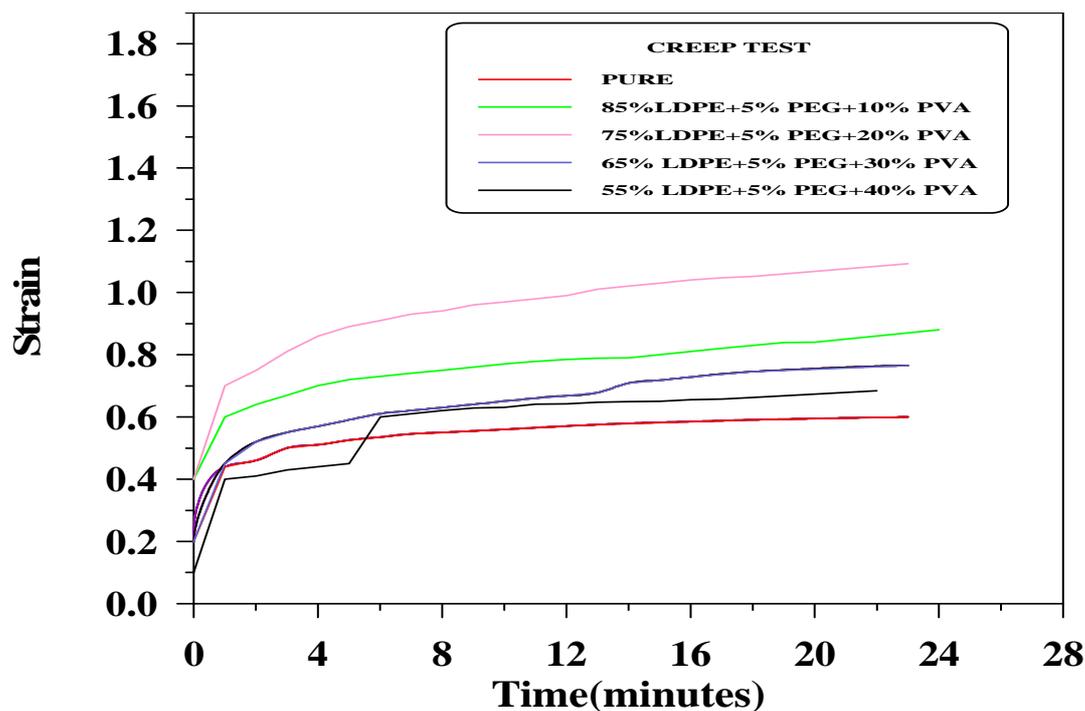


Fig. 5: Show (strain-time) curve of pure LDPE and LDPE+5% PEG with PVA in different percent.

4.3 FTIR Analysis:

Through this analysis will be found the chemical structure of LDPE/ PVA and LDPE/PEG/PVA according to bands values, recorded by Fourier transform spectroscopy (FTIR) for the pure polymers and blend prepared by method of extrusion with different composition of blends are summarized in Table (4-1), which were derived from Figure (4-13) respectively. FTIR test for low density polyethylene shows many bands such as the bands at 2921 and 2849 cm^{-1} for (C-H stretching), the band at 1465 cm^{-1} for (CH_2 bending), The band at 720 cm^{-1} for (CH_2 rocking).

The blends from (LDPE/PVA) and (LDPE/PEG/PVA), the bands at 2921 cm^{-1} and 2849 cm^{-1} shifted to 2920 cm^{-1} and 2850 cm^{-1} respectively for (LDPE/PVA), 2920 cm^{-1} and 2846 cm^{-1} respectively for blends with (PEG).

the band at 1465 cm^{-1} appear is the same for (LDPE/PVA), 1467 cm^{-1} for the blends with (PEG), the band at 720 cm^{-1} shifted to 721 cm^{-1} for (LDPE/PVA), 723 cm^{-1} with (PEG), as show in figure (4-13).

The results from FTIR test not show any reaction between them, this due to nobonds appear, while the results show some bonds shifting this due to physical interaction.

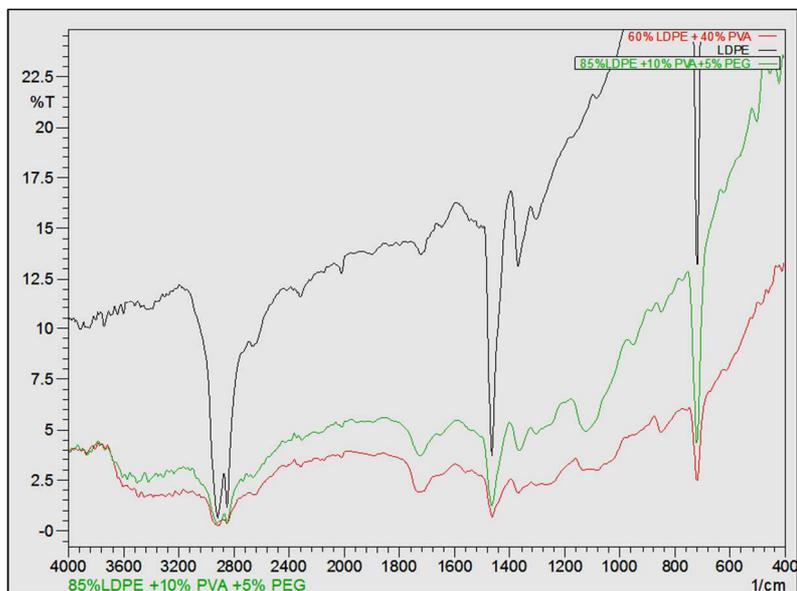


Fig. 6: Show FTIR for LDPE and LDPE/PVA and LDPE/PEG /PVA

Table 2: The absorption bands of the IR spectrum characteristic of LDPE and blends

Type of bond	Stander LDPE ^[9,10]	Exp. LDPE	LDPE+PVA	LDPE+PEG+PV A)
CH ₂ stretching	2914	2921	2920	2920
	2846	2849	2852	2846
CH ₂ bending	1474	1465	1465	1467
CH ₂ rocking	720-724	720	721	723

4.4 Density test:

The effect of addition of PVA to the LDPE is shown in figure 7, this figures indicates that the density increase with increasing PVA percent that is related to the PVA fill the void which has higher density values.

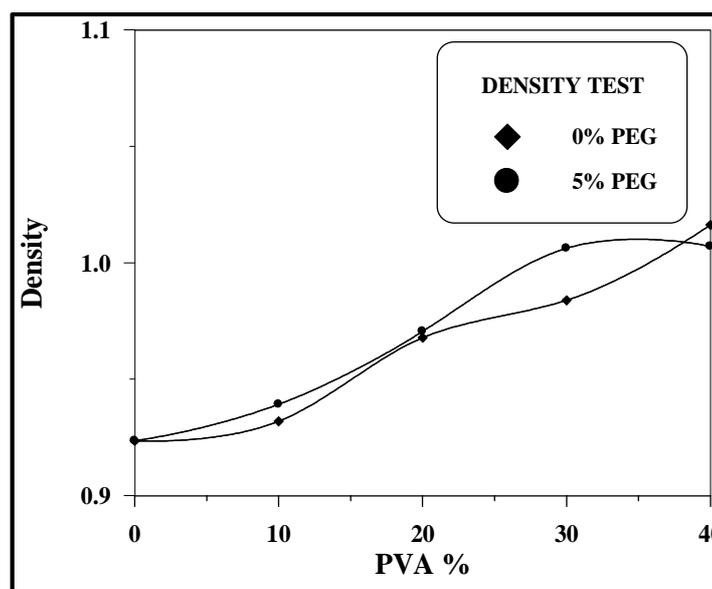


Fig. 7: Effect of PVA on LDPE density

4.5 Morphology test:

The influence of degradation time on the biodegradability of the blend sample was obviously seen in the digital microscope (200X).

Before burial test in soil, the smooth surface of LDPE and the blend samples was observed. Inconstant, the sample after burial in soil were rough and had a lot of small pitting on the surface, whereas the LDPE sample remained unchanged. The existence of pitting can be noticed on the surface of the sample in soil. However, the polymer samples gross morphology was observed to be changed physically, for example the surface pitting over the degradation period, the increased PVA constant resulted in higher surface pitting of the sample due to increased degradation. The change in physical appearance of the sample in the soil could be considered as evidence of biodegradation of this polymer in the landfills or natural environment. The result indicate that the incorporation of hydrophobic LDPE with hydrophilic enhance the hydrophobicity and degradability of the overall polymer as show in figure 8.

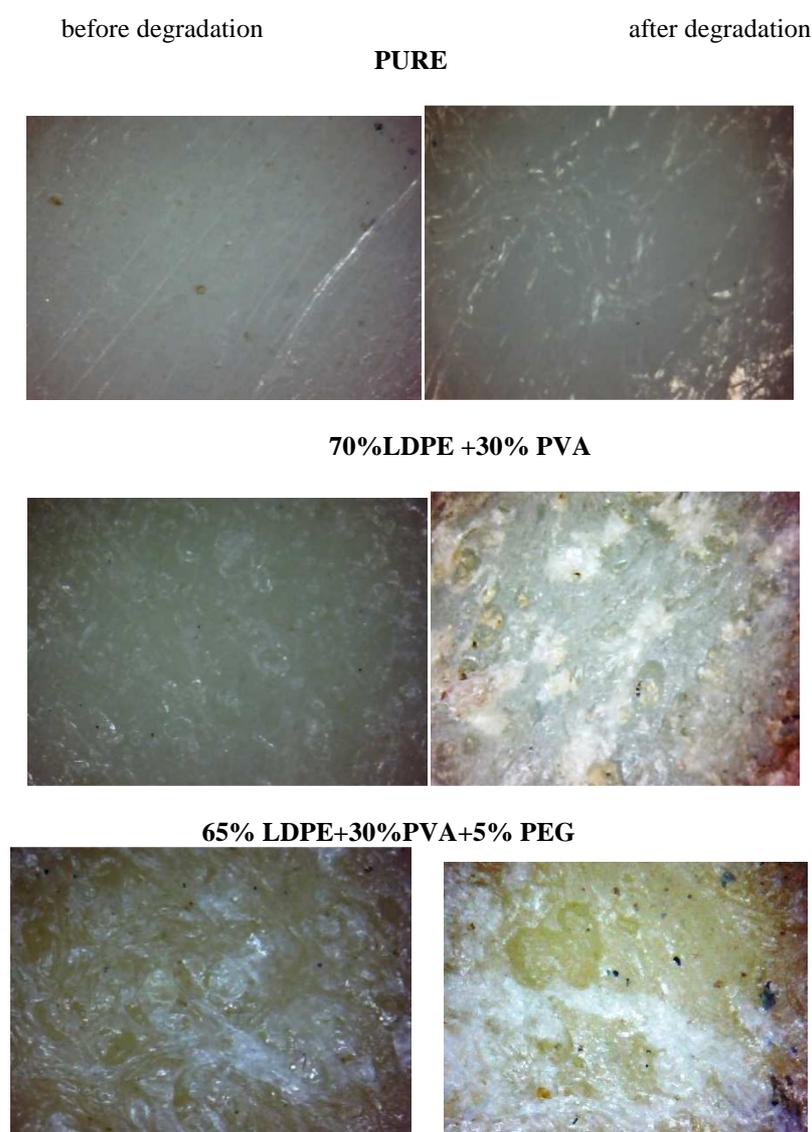


Fig. 8: Surface morphology of the LDPE polymer with different percentages of PVA.

4.6 Scan Electron Microscope (SEM) Test:

Figures 9 to figure 11 show the SEM images of the samples. Figure 10 show PVA formed agglomerates and show without any adherences with the matrix and can distinguished it clearly in fracture, so the PVA unsuitable for mechanical adhesion this is the reason for a weak mechanical properties of the blends.

In case the addition PEG to the same blends as show in figure 11, addition PEG led to increase size the formed agglomerate this agree with the results of mechanical properties which are obtained previously.

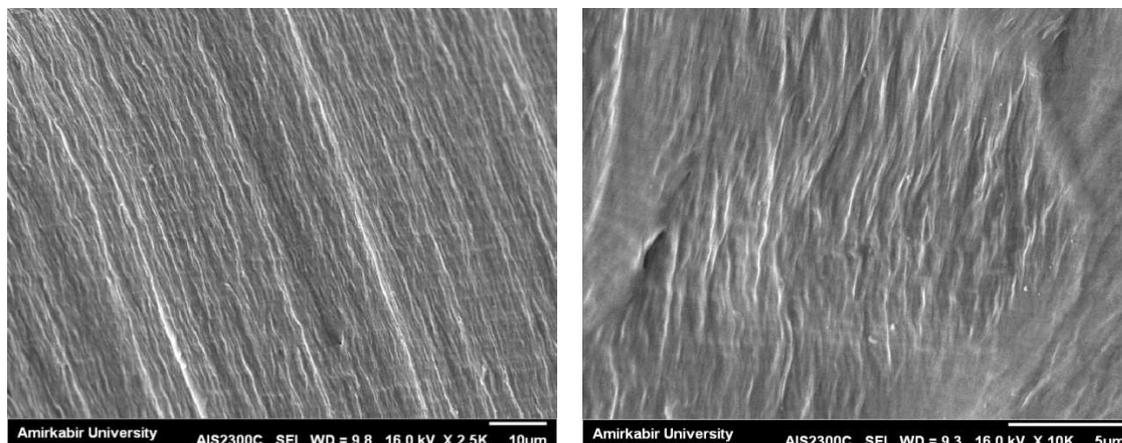


Fig. 9: SEM Images of pure LDPE a , b Respectively.

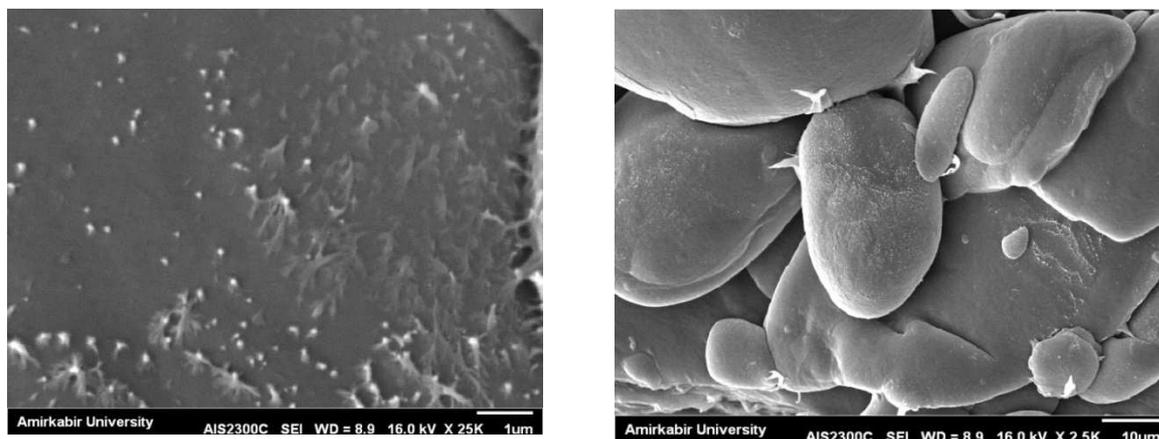


Fig. 10: SEM Images of 70%LDPE+30%PVA a , b Respectively.

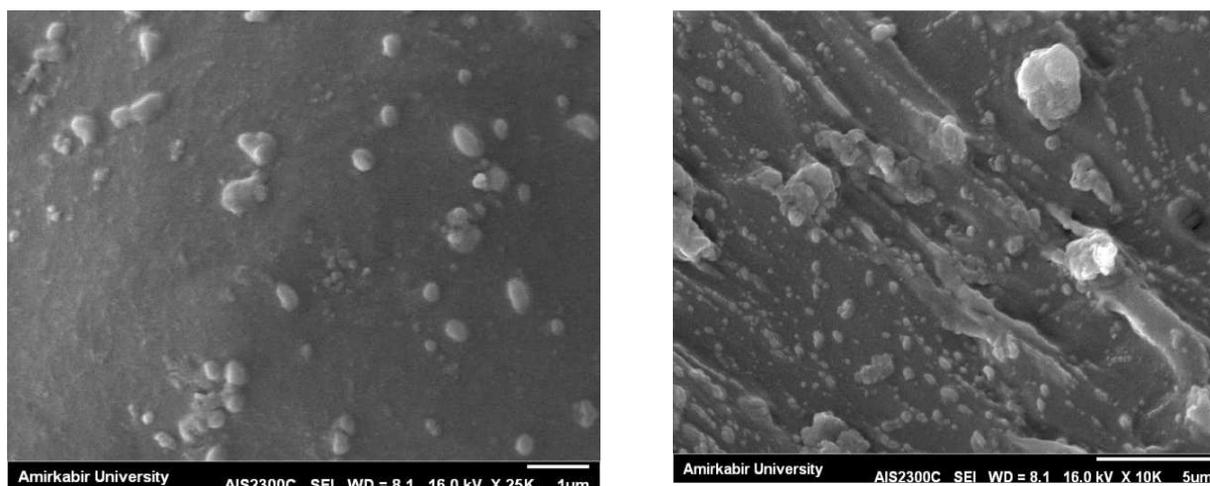


Fig. 11: SEM Images of 65%LDPE+30%PVA+5%PEG a , b Respectively.

4.7 Degradation tests:

4.6.1 Soil burial test

Figure 12 shows weight loss(%) of LDPE / PVA blends against degradation time in soil environment.

Results showed that Pure LDPE Sample showed no weight loss and no surface deterioration in soil environments within the 100-day study period, On the other hand the weight losses increase with the amount of PVA increase. These results reveal that higher PVA content enhances the degradation kinetics and thus increases mass loss. This could be due to the hydrophilic nature of PVA. The higher PVA content in the blend, the higher the moisture content that renders faster degradation.

When adding 5% PEG of samples above observe increase the proportion of weight loss when placed in soil for 100 days with increase PVA percent added to LDPE due to the nature of the PEG hydrophilic and ability on the biodegradation as shown in Figure13

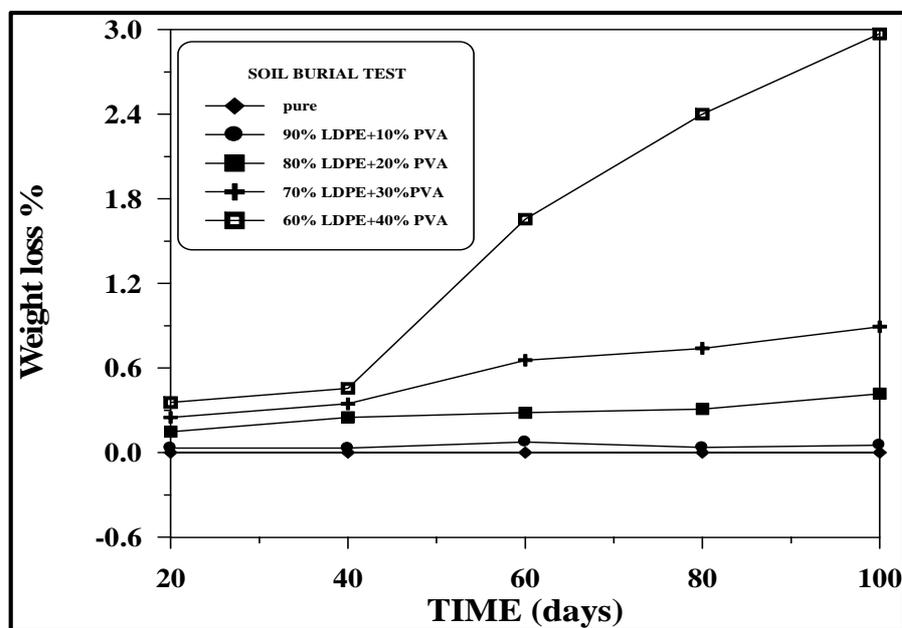


Fig. 12: show biodegradability of LDPE/PVA blend in soil environment

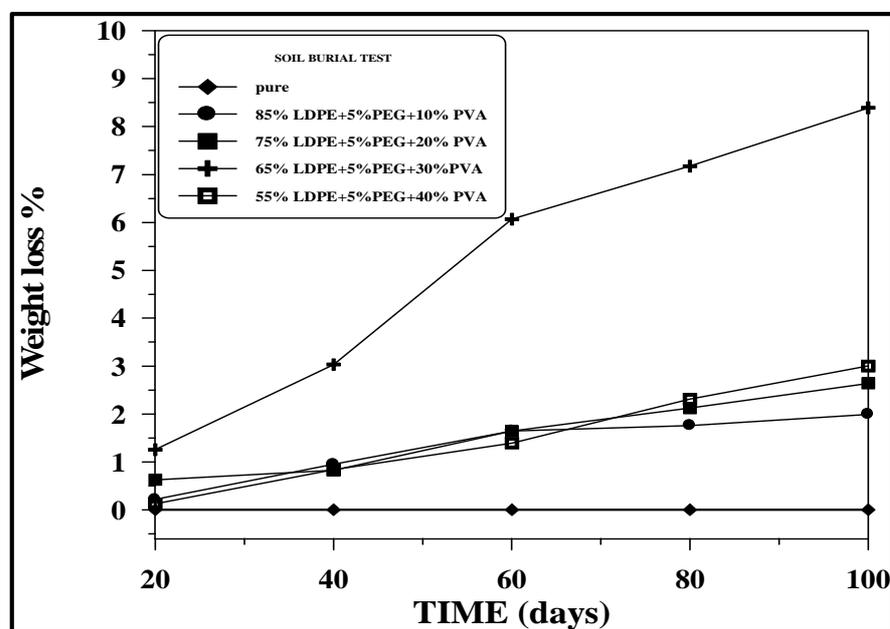


Fig. 13: show of biodegradability of LDPE/PVA /PEG in soil environment

Conclusions:

1. The tensile properties such as tensile strength and elongation at break of the LDPE/PVA blends decrease with increase PVA percent, and the addition 5% PEG of the blends lead to more decrease tensile properties, while young's modulus increase.

2. The soil burial test of LDPE/PVA blend showed that, low density polyethylene sample showed no weight loss and no surface deterioration in soil environments within the 100-day study period. On the other hand the weight losses increase with the amount of polyvinyl alcohol increase.

3. Adding 5% PEG of samples above observe increase the proportion of weight loss when placed in soil for 100 days with increase PVA percent added to LDPE.

4. The physical appearance studies of LDPE /PVA blend after burial test in soil showed that all blend films was obviously changed after 100-day study period, whereas the pure LDPE samples remained unchanged.

5. The morphology test result showed that voids and pitting are formed after burial in soil increased with increasing the time which indicates clearly the prevalence of degradation process.

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