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Research Article

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Mechanical and Thermal Properties of Olive Solid Waste/ Recycled Low Density Polyethylene Blends

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Abstract Low density polyethylene (LDPE) and linear low density polyethylene (LLDPE) are widely used in plastic film industries, causing a large quantity of waste with unpredictable mechanical and thermal properties. Olive solid waste, an abundant material usually thrown into land causing harms to environment, was mixed with recycled plastic films and used as a modifying filler. The mixture was blended by a homemade single screw extruder operating at constant screw speed and zone temperatures. Three olive solid waste particle sizes, less than 150 µm, 150-250 µm and larger than 250 µm, were used. The effects of olive solid waste contents from 0 to 25 wt.% on mechanical and thermal properties were tested, it was found that the addition of olive solid waste particles, of less than 150 µm, up to 10wt.% resulted in a noticeable improvement of elastic modulus and ultimate tensile strength. While for other particle sizes, 150-250 and larger than 250 µm, the optimum values of both ultimate tensile strength and elastic modulus were found at 2.5 wt.% olive solid waste content. Besides that it was found that an addition of these particles enhances the degree of crystalinity of the polyethylene blend and improves the compatibility between both polymers used to manufacture plastic films (LDPE and LLDPE).

Keywords Plastic films, Olive solid waste, Elastic modulus, Ultimate tensile strength, Degree of crystallinity

1. Introduction

Recently, synthetic polymers such as polyolefins are used in many applications because they have many advantages such as low cost, light weight, moderate mechanical properties, and ease of processing, but they are not biodegradable materials which causes environmental pollution. In order to solve this problem, scientific research was directed towards recycling these polymers to produce new materials that can be used in several applications such as floor panels, interior house decorate parts. Unfortunately, the properties of recycled polymers such as mechanical properties are decreased during the processing stage, so the best method to improve these properties is to reinforce recycled polymers with a suitable reinforcement. Both organic and inorganic reinforcements can be used to improve the mechanical and thermal properties of recycled synthetic polymers.

Different types of reinforcements such as sawdust, carburized solid olive waste, potato starch and jute fibers were used to improve the tensile and flexural properties of polyethylene, the degree of improvement was found to be affected by several factors such as reinforcement type, its content, its size, and the compatibility between the polymeric matrix and reinforcement [1-6].

organic reinforcement are biodegradable materials that are derived from renewable resources, and they have several advantages such as low cost and light weight, so the blending of polyethylene with organic reinforcements has received a considerable attention. Different types of these reinforcements, such as sawdust [7, 8], carbon particles [9], corn starch [10], potato starch [1], rice [2], jute fibers [3], linen yarn products waste

[4], olive oil waste [11], chars obtained by co-pyrolysis of olive pomace [12] were used to improve the properties of both low density and high density polyethylene (LDPE and HDPE).
Due to difference in hydrophobicity between polyethylene and reinforcements, suitable compatibilizers such as PE-grafted-maleic anhydride (PE-g-MA) and PP-grafted-maleic anhydride (PP-g-MA) were used [13,14].Abdul Majid *et al* studied the effect of addition of PE-g-MA on the properties of LDPE-thermoplastic sago starch reinforced kenaf fiber composites. The tensile strength, elastic modulus and water absorption were found to be improved by the addition of the compatibilizer [13]. The effect of PP-g-MA on the properties of recycled HDPE- coconut, eucalyptus, or pine fiber composites was investigated. The use of compatibilizer improved the strength properties as well as the water absorption of the composites over those without compatibilizer [14].
It is obvious that there is tremendous amount of olive solid waste present in Palestine and Mediterranean area. Considering the available recycled LDPE and olive solid waste amounts, manufacturing of polymer based composites could be a viable option for plastic industry in Palestine. In this study, the effects of the olive solid waste content and particle size on mechanical as well as thermal properties of the final blend (LDPE/olive solid waste) were investigated.

2. Materials and Methods

2.1. Materials

The material used in this work was recycled low density polyethylene (LDPE) and linear low density polyethylene (LLDPE) blend in the form of granules obtained from local plastic film industries. Olive solid waste (OSW) which is an abundant material produced as a by-product from olive squeezing process was used as a modifying and filling additive in the form of particles at different sizes (<150µm, 150-250 µm and >250 µm) the olive solid waste was dried at 120°C for 4 hours to remove their moisture content.

2.2. Processing

2.2.1. Extrusion Process

Low density polyethylene and olive solid waste (OSW) were mixed together in different weight percent (2.5, 5, 10, 15, 20 and 25 wt.%) by the use of simple tumbling device. The blends were introduced into the feed zone of home single screw extruder which has length to diameter (L/D) ratio of 20. The prepared samples were extruded at 160°C for all zones (feed, compression, metering and die) and at a screw speed of 17 round per minute (rpm), then emerged from a capillary die of 10 mm diameters and cooled in a water path to room temperature.

2.2.2. Compression Molding

A homemade thermal press was used to prepare the samples sheets, it consists of two metallic palates which can be heated electrically, the pressure was applied by using the air pressure using a compressor. The thermal press is equipped with a water cooling system. The extruded samples were pressed into 1.5 mm thick sheets using a 8×10 frame model by the use of homemade thermal press molding apparatus operated at a pressure of 5-6 bar (gauge). The samples were pressed at 200°C for 15 minutes, and then the sheets were removed after cooling the press through installed cooling water system.

2.3. Characterization of Composite Materials

2.3.1. Tensile Test

Tensile test was carried out by using universal testing machine (Gunt Hamburg apparatus WP 310 machine)at a constant speed of 4 mm/minute. For each composite, five specimens of 130 mm gauge length, 10 mm width and 1.5mm thickness were tested. The test was carried out according to standard test method for tensile properties of plastic ASTM D638-14.

2.3.2. Thermal Test (DSC)

Thermal properties of recycled polyethylene blend-olive solid waste samples were tested using a differential scanning calorimeter (DSC)model Pyrix-6, PerkinElmer Corporation, U.K. For each sample, 5 to 8 mg was placed into a sealed aluminum pan. The same temperature profile (from room temperature to 200 ^{0}C), heated at 10°C/ minute was applied to all samples.Melting temperature and heat of fusion of the samples were obtained from the maximum peak and area under the peak, respectively, where the last one is essential to estimate the percentage of crystalline regions.DSC test was done with regard to ASTM D3418-15 (Standard Test Method for Transition Temperatures and Enthalpies of Fusion and Crystallization of Polymers by Differential Scanning Calorimetry).

3. Results and Discussion

3.1 Tensile Properties

The effects of olive solid waste content and particle size on the tensile properties of polyethylene blend-olive solid waste composites are shown in Tables (1-3).

Table 1: Mechanical properties of composite materials based on olive solid waste (<150 µm) / recycled polyethylene blend

OSW content (wt.%)	Ultimate tensile strength (MPa)	Elastic modulus (MPa)	Ductility (%EL)
0	08.75 ± 0.42	311.5 ± 37.2	277.7 ± 29.7
2.5	10.18 ± 0.62	559.2 ± 37.3	177.4 ± 16.4
5	10.48 ± 0.91	588.5 ± 14.2	101.2 ± 6.4
10	10.65 ± 0.35	608.2 ± 34.2	100.2 ± 5.4
15	10.09 ± 0.92	557.5 ± 67.0	97.6 ± 5.6
20	08.90 ± 0.31	332.2 ± 65.3	89.1 ± 7.4
25	07.71 ± 0.46	238.1 ± 60.3	64.8 ± 6.4

Table 2: Mechanical properties of composite materials based on olive solid waste (150-250 µm) / recycled polyethylene blend

OSW content (wt.%)	Ultimate tensile (MPa)	Elastic modulus (MPa)	Ductility (%EL)
0	8.75 ± 0.42	311.5 ± 37.2	277.7 ± 29.7
2.5	10.0 ± 0.50	479.6 ± 18.0	105.2 ± 2.0
5	9.79 ± 0.44	479.4 ± 12.9	93.40 ± 6.6
10	8.60 ± 0.42	413.6 ± 66.0	89.80 ± 17.9
15	8.27 ± 0.32	375.5 ± 61.6	81.50 ± 13.4
20	8.17 ± 0.70	323.8 ± 58.0	60.00 ± 6.4
25	7.49 ± 0.38	272.3 ± 60.2	50.00 ± 5.1

Table 3: Mechanical properties of composite materials based on olive solid waste (>250 µm) / recycled polyethylene blend

OSW content (wt.%)	Ultimate tensile strength (MPa)	Elastic modulus (MPa)	Ductility (%EL)
0	8.75 ± 0.42	311.5 ± 37.2	277.7 ± 29.7
2.5	9.20 ± 0.54	435.0 ± 44.2	91.47 ± 11.8
5	8.22 ± 0.40	424.8 ± 57.5	85.86 ± 16.3
10	8.15 ± 1.01	312.5 ± 71.5	76.38 ± 10.9
15	8.03 ± 0.70	299.2 ± 55.4	71.62 ± 3.3
20	7.96 ± 0.67	273.2 ± 30.0	56.67 ± 6.4
25	7.06 ± 0.86	187.7 ± 32.0	47.27 ± 11.8

It was found that olive solid wastes with different particle sizes have a clear effect on tensile properties of recycled low density polyethylene blend. An addition of these particles increases both the ultimate tensile strength and elastic modulus up to a certain weight percent after which the properties start to decay, this may be related to the formation of voids at high olive solid waste content. This decrease is dependent on the size of the particles, olive solid waste particles having a size less than 150µm have better results and can improve both elastic modulus and ultimate strength up to values of 608.2±34.2 and 10.65±0.35MPa, respectively, with an addition of 10wt.% of olive solid wastes as shown in Figures (1-2), but this increase can be noticed only up to 2.5wt.% of an addition of olive solid waste for the other particle sizes (150-250 µm and >250 µm) as shown in Figures (1-2). This decrease may be related to the larger volume of voids which may be formed within the blend or due to smaller surface area of these particles which adversely affects the interfacial adhesion between the particles and the polymeric matrix, it is worthy here to point that other researchers had found the same behavior of polymers with solid additives where fine particles have a better effect than larger ones [2,15].

The increase in tensile properties was significant for elastic modulus which is an important mechanical property for engineering polymers and a slight increase in ultimate tensile strength which is a maximum stress that the polymer can withstand before fracture, this increase can be related to an increase in degree of crystalinity of PE or to some cross-linking occurred due to an addition of olive solid waste which may contain some active materials such as metallic polymeric compounds (poylmerin) [16].

On the other hand, ductility of the produced composites was estimated to observe the effect of olive solid waste content on the ductility of the polymeric matrix; this effect is clear in Figure 1.It is obvious that composite materials exhibit a significant decrease in ductility at lower olive solid waste concentration and then its value decreases at lower rate. This behavior could be attributed to the formation of dense structures within the blend volume as the olive solid waste concentration increases, which could produce anisotropic final product.

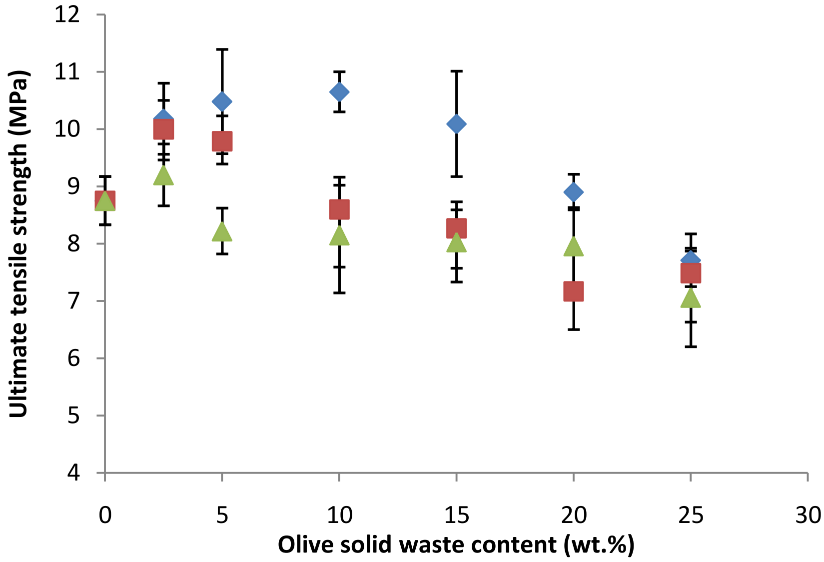

Figure 1: Ultimate tensile strength of polyethylene blend/olive solid waste based composites, where ◆, ■ and ▲ represents olive solid waste (<150 µm), olive solid waste (150-250 µm) and olive solid waste (>250 µm)

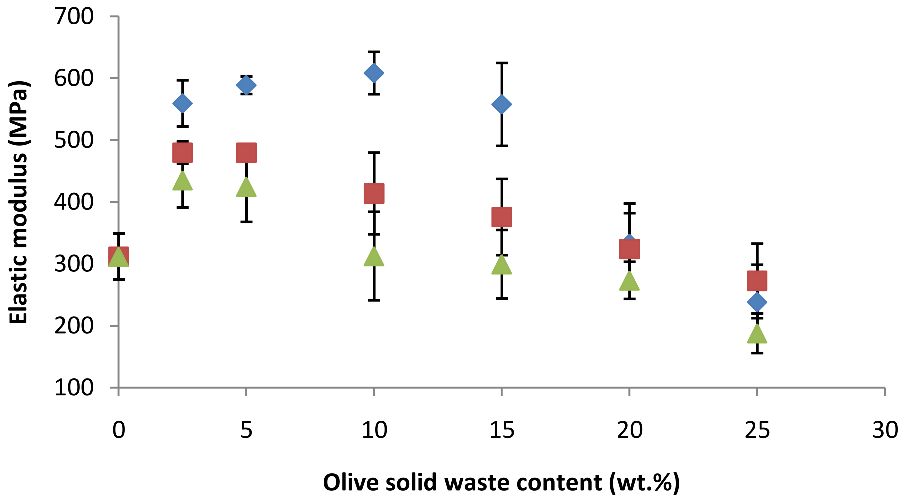

Figure 2: Elastic modulus of polyethylene blend/olive solid waste based composites, where ◆, ■ and ▲ represents olive solid waste (<150 µm), olive solid waste (150-250 µm) and olive solid waste (>250 µm)

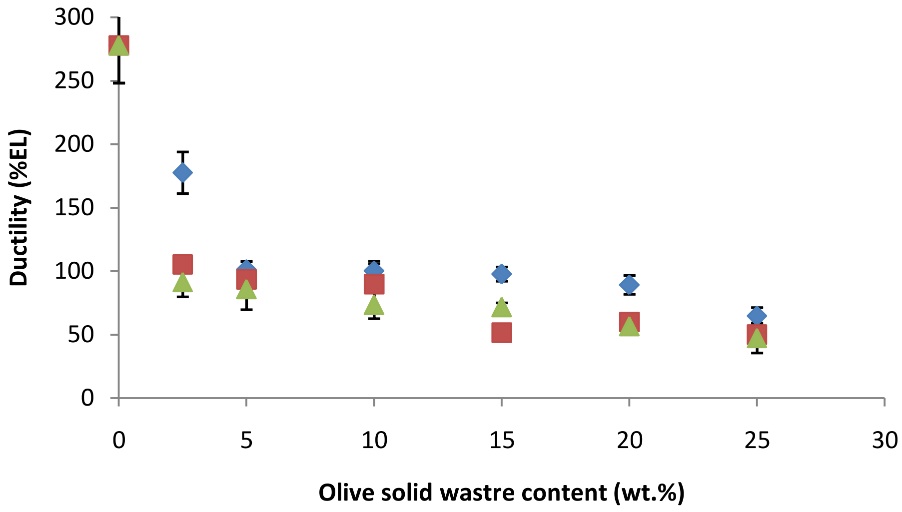

Figure 3: ductility of polyethylene blend/olive solid waste based composites, where ◆, ■ and ▲ represents olive solid waste (<150 µm), olive solid waste (150-250 µm) and olive solid waste (>250 µm)

3.2 Thermal Properties

It is claimed that the improvement in mechanical properties is attributed to the presence of reinforcement which acts a load carrier and/or to the increase in degree of crystallinity of the polymeric matrix, therefore it is important to test the thermal properties of the produced composites.

A 5wt.% samples for three particle sizes were scanned by a DSC in the range of 80-160°C and the results are shown in Figure 4.

It is clear from Figure 4 that the curve of recycled LDPE (curve a) exhibits two melting peaks at 110.5 and 122.2°C which means that it composed from two different immiscible polymer blend, which are LDPE and LLDPE [9]. In plastic film industries LLDPE blended in minor concentration with LDPE to improve productivity and increase throughputs [17] where they are immiscible at these concentrations [18-20].

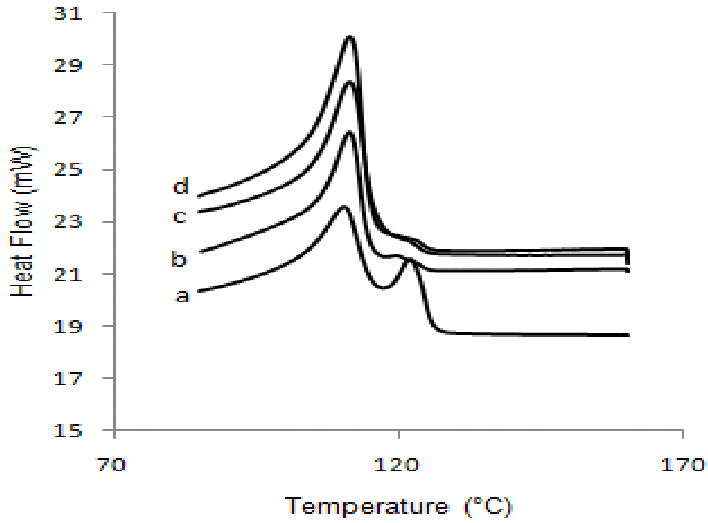

Figure 4: DSC results for a) recycled polyethylene blend, b) 5wt.% olive solid waste (<150µm)/recycled polyethylene blend,c) 5wt.% olive solid waste (150-250µm)/recycled polyethylene blend and d) 5w.t% olive solid waste (>250 µm)/recycled polyethylene blend

An addition of olive solid waste partially eliminates the second peak as shown in Figure 4 curves b, c and d which means that the blend is partially miscible, this may be due to phenolic or polar compounds existed in the olive solid waste [17] where they may enhance the miscibility between LDPE and LLDPE by the formation of functional groups and acting as a compatibilizer for both existing polymers. On the other hand, it is worthy to find the area under the melting peaks to find heat of fusion which is an indication on the percentage of crystalline regions where the last can be calculated from the following equation:

$$x_c = \frac{\Delta H_f}{\Delta H_f^0 X \varphi} X 100\%$$

Where X_C is the percentage of crystalline regions, ΔH_f is heat of fusion of the sample; ΔH°_f is heat of fusion for 100% crystalline polyethylene which is found to be 290J/g [21] and φ is weight percent of polyethylene in the blend. Melting points, heat of fusion and percent of crystalline regions for different olive solid waste particles at 5wt.% are shown in Table (4).

Table 4: Thermal analysis data of 5wt.% olive solid waste/ recycled low density polyethylene blend

Particle sizes (µm)	**Melting temperature (°C)**	**ΔH^0_f (J/g)**	**Xc (%)**
-	110.5, 122.2	79.2	27.3
<150	111.5	96.7	35.1
150-250	111.6	86.5	31.4
>250	111.7	84.0	30.4

It is obvious that an addition of fine particles has significant effect on the degree of crystallinity, but this effect is inversely proportional with the particle size, the degree of crystallinity increased from 27.3% for recycled low density polyethylene blend to 35.1% for recycled low density polyethylene blend containing smaller particles (<150µm), but it increased to 31.4% and 30.5% for recycled low density polyethylene blend containing particles having a size of 150-250 µm and >250 µm, respectively.

The degree of crystallinity affects the tensile strength and modulus of elasticity of the produced composites as shown in Tables (1-3) and Figures (1-2). It can be seen from the tables that the tensile strength and modulus of elasticity of composites containing 5wt.% are inversely proportional with the particle size, this may be related to the several reasons, as mentioned above, and to the increase in the degree of crystallinity of PE.

4. Conclusion

From the previous work, it was concluded that olive solid waste improved both the mechanical and thermal properties of recycled plastic films. For mechanical properties, the improvements were observed for both ultimate tensile strength and elastic modulus, they were increased by the addition of the olive solid waste up to a certain level, then they stated to decay due to the formation of voids and loss in the ability of the polymeric matrix to impregnate and wet the solid particles, the maximum observed points for tensile properties were varied according to the size of the added olive solid waste particles, fine particles had better effect with 10 wt.% maximum weight percent and that was due to their large surface area and smaller sizes of voids created within the produced samples. While other sizes showed a maximum properties at lower level with less amount of olive solid waste which was about 2.5wt.%. On other hand, these improvements were greatly related to the degree of crystalinity which was enhanced by the addition of olive solid waste, and also the addition of solid waste particles enhanced the compatibility between LDPE and LLDPE to become partially miscible blend.

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