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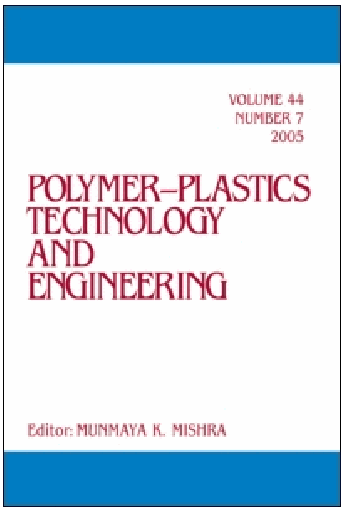

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Improvements of the Tensile Properties of Recycled High Density Polyethylene (HDPE) by the Use of Carbonized Olive Solid Waste

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Recycling of plastic poses several concerns to manufacturers. The most important concern is the unpredictable of their mechanical properties (modulus of elasticity, tensile strength and ductility). Olive solid waste, an abundant material usually thrown into land causing harms to environment was mixed with HDPE plastic and used as a filling material. The mixture was fed to a house made extruder operating at different speed and temperature. Two carbon particle sizes range (less than 150 μm and 180–250 μm) were used. The effect of carbon contents from 0 to 10% wt/wt and operating conditions were tested on the mechanical properties of the recycled HDPE plastic. It was found that up to 5% wt/wt carbon of less than 150 μm resulted in a noticeable improvement of modulus of elasticity and tensile strength. The optimum value of modulus at carbon particle size 180–250 μm was found at 2.5 olive solid carbon content. Increasing screw speed was found to increase tensile modulus and strength of used plastic. This was related to melt viscosity and reduction in particle size. An increase in processing temperature was found to improve tensile properties up to certain point where degradation of polymeric matrix begins to occur and therefore tensile properties deteriorate.

Keywords Carbon particle sizes; Extruder operating variables; HDPE; Olive solid waste; Tensile properties

INTRODUCTION

The environmental problem nowadays is a factor of extreme importance in the industrial world, particularly in the case of plastic processing companies. Plastics have become one of the materials with the greatest growth in terms of consumption and in the amount of generated waste[1]. Efforts are mainly focused on the reduction and recycling of wastes generated during transformation processes and also after product end use. Although the recycling capacity for plastics has been progressively increased, the fraction of plastics that end up in a landfill is still very significant[2].

The recycling of plastic wastes is not a recent problem for plastic users and producers. Since long, industrial scraps are recycled within the production cycle itself or recovered as lower-grade materials and re-fabricated into new products. Recycled HDPE can be used in a growing number of potential applications such as boxes or pellets, whenever the thermal, mechanical and impact properties of the recycled polymer are close to the ones of virgin material. This is one of the goals for the general use of recycled polymers[3]. There is a great interest in finding new possibilities for the use of post consumer plastics as new product[4–7].

During the successive processing steps, irreversible thermo-oxidative degradation in the form of chain scission, cross-linking or elimination of substituent and formation of double bonds can be produced yielding recycled materials whose properties are usually lower than the original ones[8]. To avoid degradation, stabilizers and antioxidants are added contributing to the preservation of the original properties[9–14]. However, their introduction normally leads to an increase in the cost of the material. To improve the processing and retain the mechanical properties, recycling strategies such as melt-blending with or without compatibilizing agents have been used[14–16]. Blends of HDPE with other polyolefin like PP, LDPE or LLDPE have attracted researchers' attention because they are low-cost and found to improve the mechanical properties of an HDPE/PS blend by compatibilization and incorporation of $CaCO_3$[17]. The incorporation of $CaCO_3$ significantly enhanced the stiffness but lowers the impact resistance of recycled HDPE.

Another possibility of obtaining low-cost recycled HDPE based polymers is the addition of particulate inorganic additives to the matrix. Introduction of fillers has the effect of reducing the cost and enhancing some of the mechanical properties like the Young's modulus or the yield strength, but on the contrary other important properties like toughness or the impact strength are usually degraded. The most common filler for HDPE is calcium carbonate[18,19], but others like mica, glass beads[20], talc[21] and clays[22] have been successfully used. Moreover, using a small particle size (2–10 μm) the talc nucleation activity is

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enhanced and the possible impact damage due to stress concentration around particles is reduced and lead to a significant change in properties.

HDPE is one of the most used polymers in the Palestinian market Post consumer HDPE from bottles is an interesting source of recycled material because, on one hand, it can not be used again in alimentary applications and, on the other hand, its high melting viscosity makes direct transformation via injection molding very difficult. It is found that the consumed plastics are thrown away improperly causing damage to the environment.

Although recycling used plastic is an option to minimize plastic waste, it was not the case in the Palestinian industry as recycling of plastic poses several concerns to manufacturers such as: (a) lack of homogeneity of the material; (b) lack of purity; (c) unpredictable in their properties and (d) may contain harmful and dangerous materials.

On short, these reasons resulted in a lack of trust by the producers about the mechanical tensile strength and modulus of elasticity and morphological properties of the recycled plastic. Therefore, it is the main aim of this work to overcome these concerns by enhancing the mechanical properties of recycled plastics using suitable, abundant material as filler. Such filler has two important characteristics:

First, it will improve the mechanical properties of the recycled plastics, mainly the Young's Modulus and the stress at break without sufficiently loss in the impact toughness. Second, it will encourage plastic producers to recycle more plastics as it will cut down the cost of manufacturing plastic containers from virgin plastic imported from the near market. The still opened question is what kind of abundant material we can use?

Olive solid waste is an abundant material with an annual production of more than a hundred thousand tones [23]. Because of its physical properties (low density and the rigidity), it can be used as subsidiary filler in plastic production compared with other additives such as wood fibers that affects mechanical properties such as flexural strength and resilience[24].

The feasibility of the suitability of using olive solid waste as a filer in plastic production had not been studied before and deserves more research; part of this long term research is the outcome of this research. The novelty of this work is that abundant cheap olive waste is used as a filler to improve the mechanical properties of recycled plastic.

EXPERIMENTAL WORK

The methodology of this work consists of choosing the raw materials (recycled HDPE and abundant materials), processing of the raw materials and finally testing the physical properties of the modified plastics.

Materials

1. High Density Polyethylene (HDPE)—Crushed high density polyethylene obtained from trimmings or unsuitable detergent bottles was selected to be modified. This choice was due to its popularity in detergent industry in Palestinian market, it was in the form of flakes.
2. Olive solid waste—Olive solid waste, which is an abundant material produced in large quantities in Palestine. It was selected to be the modifier and filler for HDPE after appropriate treatment.

Processing

Treatment of Olive Solid Waste. Olive solid waste was cleaned from the residual oil by the mean of Soxhlet method, and then dried in the oven at 120°C for 2–3 hours. Olive solid waste has satisfactory physical and chemical properties that allow it to be utilized in water cleaning. Take as an example its moderate internal surface area (a physical property), which allows it to be utilized in water treatment[23].

Olive solid waste has a high quantity of volatile matter, which when carbonized in absence of oxygen at 600°C for 1 hour, results in a carbon porous structure. Carbon particles were grinded by a mill into different sizes, which were sieved and separated. Particles less than 150 μm and particle of 180–250 μm were chosen to be blended with flakes of HDPE.

Sample Preparation. HDPE flakes were mixed by simple tumbling with olive solid waste carbon to prepare different samples with carbon content of 2.5, 5, 7 and 10%. For each carbon content, two samples of different particle size were prepared. One with particle less than or equal 150 μm and other of 180–250 μm.

The blends were introduced into the feed zone of a home-made single screw extruder had L/D ratio of 20 with three distinct zones (feed, compression, and metering). The flight depth in the metering zone was 2.25 mm, helix angle 17.7°, which could be operated at different rotating speeds varied from about 7 to 28 rpm. The extruder was attached with a capillary die of 10 mm in diameter. The prepared samples were extruded at the same temperature 220°C and the same screw speed of 17 rpm. The samples emerged from the die were cooled to room temperature by a water path.

A 5 wt% sample prepared from particle of size $\leq 150\,\mu m$ was processed at different screw speeds (7, 17, 28) and at different temperatures (200, 220 and 240°C) to examine the effect of operating variables on the tensile properties of the blend.

Testing

Tensile Test. Tensile test was carried out by using Gunt Hamburg apparatus WP 310 machine at constant speed of 5 mm/min and at room temperature. For each sample, five specimens were tested. The samples were 130 mm gauge length and 6 mm gauge diameter. Tensile Test provides values of the tensile strength, modulus of elasticity and ductility as percent elongation.

RESULTS AND DISCUSSIONS

Effect of Carbon Weight Percent and Particle Size

Two carbon olive solid waste particles sizes (less than 150 μm and 180–250 μm sizes range) were used at different blend of carbon-HDPE (0–10%), where 0% means 100% recycled HDPE. The effect of carbon blend at same particle size on tensile properties was tested. Results for carbon particle size ranges of less than 150 μm and 180–250 μm are listed in Tables 1 and 2, respectively.

It was found that weight percent and size of carbon particles have clear effects on the mechanical properties of recycled HDPE. Effect of carbon particle size of ≤ 150 μm and 180–250 μm on modulus of elasticity, tensile strength and ductility are shown in Figures 1–6.

It was found that increasing the carbon content increases both the tensile strength and modulus of recycled HDPE. When carbon content is raised above a certain value both tensile strength and modulus decrease. The increase of tensile strength and modulus is function of carbon particle size. When carbon particle size of less than 150 μm, the relative increase in the modulus of elasticity at 2.5% carbon content is 13.7% increases to 57% when particle content increases to 5% (Figure 1).

TABLE 1
Mechanical properties of recycled HDPE blended with carbon particles size of less than 150 μm at different carbon content

Carbon (wt%)	Modulus of elasticity (Mpa)	Tensile strength (Mpa)	Ductility (%EL)
0.000	860.0 ± 90.00	26.00 ± 2.000	22.10 ± 1.810
2.500	978.3 ± 91.47	28.12 ± 3.500	17.10 ± 2.200
5.000	1352 ± 139.6	30.68 ± 1.500	5.300 ± 0.640
7.500	409.0 ± 60.00	13.39 ± 3.000	7.830 ± 1.760
10.00	348.4 ± 43.00	11.58 ± 1.500	8.628 ± 1.700

TABLE 2
Mechanical properties of recycled HDPE blended with carbon particles size of 180–250 μm at different carbon content

Carbon (wt%)	Modulus of elasticity (Mpa)	Tensile strength (Mpa)	Ductility (%EL)
0.000	860.0 ± 90.00	26.00 ± 2.000	22.10 ± 1.810
2.500	936.3 ± 43.20	27.16 ± 2.200	13.22 ± 1.100
5.000	610.6 ± 40.10	17.18 ± 1.800	14.25 ± 0.450
7.500	385.2 ± 45.00	14.29 ± 2.000	15.10 ± 0.910
10.00	336.5 ± 20.21	10.81 ± 1.500	15.98 ± 1.040

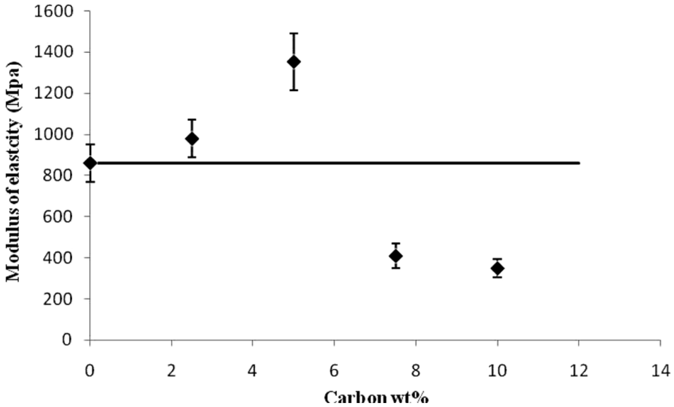

FIG. 1. Effect of carbon particles (≤150 μm) on modulus of recycled HDPE.

This is not the case with carbon particle size of 180–250 μm as the maximum relative increase in the modulus of elasticity is 8.8% at 2.5% carbon content (Figure 2). Above this carbon content, the relative modulus of elasticity is 29% less than the modulus of elasticity of 100% recycled HDPE. All values are compared to 100% recycled HDPE value shown as horizontal line. The horizontal lines in Figures 1 and 2 represent the value of modulus elasticity and tensile strength respectively at zero carbon content, i.e., samples with no olive solid waste carbon.

Same behavior was found for the measured tensile strength. The maximum relative increase in the tensile strength was found to be 17.9% at 5% carbon content of carbon particle size of less than 150 μm (Figure 3). The maximum relative increase in the tensile strength for the 180–250 μm was found 4.5% at 2.5% carbon content (Figure 4).

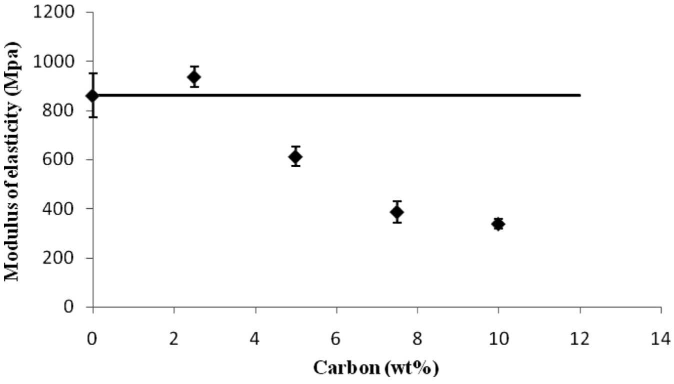

FIG. 2. Effect of carbon particles (180–250 μm) on modulus of elasticity of recycled HDPE.

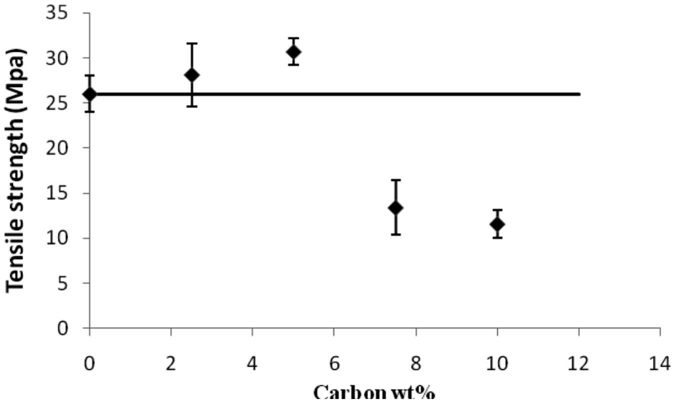

FIG. 3. Effect of carbon particles (≤150 μm) on tensile strength of recycled HDPE.

Beyond a certain weight percentage, the modulus of elasticity and tensile strength values decreased to lower values, which could be due to the formation of huge amounts of voids and loss of compatibility between carbon particles and polymeric matrix. Furthermore it can be related to the accumulation of particles at high percentages compared with lower ones, where large particles do not combine well with HDPE because of lower surface area they have.

Improvement obtained by large particles is found to be less than that obtained by fine ones and that may be as a result of the size of voids that may occur during processing. Same findings for Talc particles was reported by Sánchez-Soto et al.[1] who reported that fine talc particles improved recycled HDPE better than large particle size.

These findings suggest that the main reinforcing effect could be due to the change in the crystalline form provided by the addition of carbon particles.

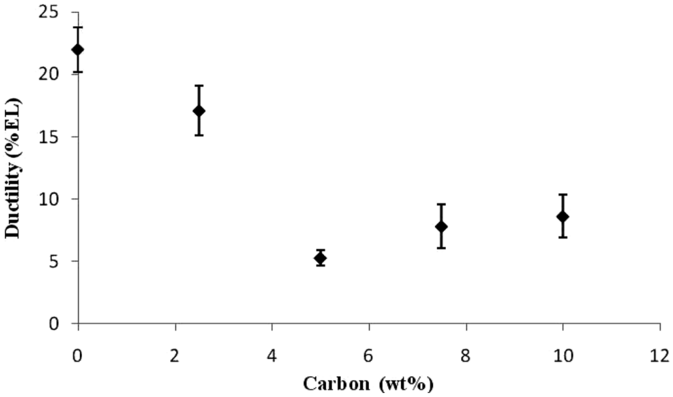

FIG. 5. Effect of carbon particles (≤150 μm) on ductility of recycled HDPE.

Ductility as percentage of elongation was found to decrease with the increase of carbon particles contents until it reaches 5 wt% when particle size is less than 150 μm. As the carbon content is increased above 5%, then the ductility is slightly increases. This is illustrated by Figure 5. In the case with carbon particle size of 180–250 μm, the ductility was found to decrease as carbon content increase until it reached 2.5% where the ductility starts to increases above this value, see Figure 6.

This ductility behavior results is in agreement with the normal relation between strength, modulus and ductility where an increase in modulus and strength leads to a decrease in ductility and vice versa.

Effect of Processing Operating Conditions

It is well known that processing techniques affect the mechanical properties of polymers; therefore it is worth to study the effect of the extruder operating conditions

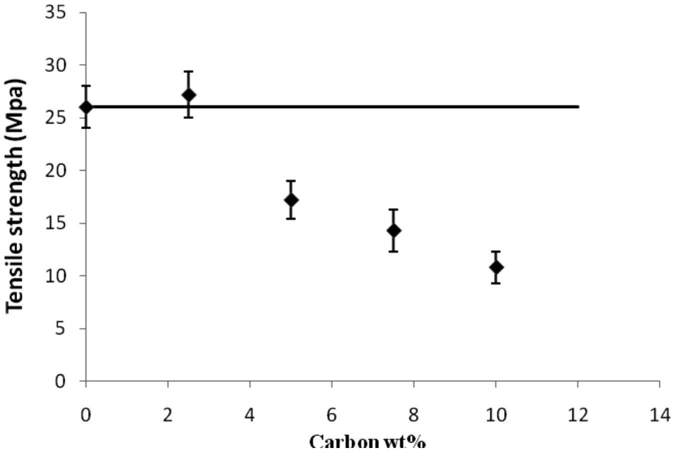

FIG. 4. Effect of carbon particles (180–250 μm) on tensile strength of recycled HDPE.

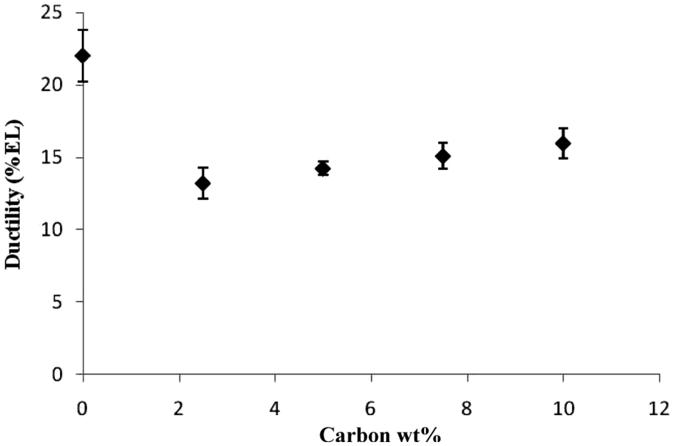

FIG. 6. Effect of carbon particles (180–250 μm) on ductility of recycled HDPE.

TABLE 3
Effect of screw speed on mechanical properties of 5 wt% carbon particles (≤150 µm)/HDPE blend processed at 220°C

Screw speed (rpm)	Modulus (Mpa)	Tensile strength (Mpa)	Ductility (%EL)
8	980.0 ± 43.0	27.00 ± 1.30	11.00 ± 0.50
17	1351 ± 139	30.68 ± 1.50	5.30 ± 0.64
28	1548.5 ± 65.0	32.50 ± 1.00	4.20 ± 0.64

on the properties of carbon particles/recycled HDPE blends.

A 5 wt% of carbon particles of less than 150 µm was blended with flakes of HDPE to be processed at different screw speeds and temperatures to estimate their effects on tensile properties as illustrated in Tables 3 and 4, respectively. Two sets of experiment were carried out to test the effect of screw speed and temperature on the mechanical properties of the recycled plastic. The first set is at constant temperature with three speed values. The second set of experiments was carried out at three temperatures and constant screw speed. All experiments were carried out at carbon content (less than 150 µm) of 5%. Screw speed values of 8, 17 and 28 rpm were chooses as they present the minimum and maximum operating speed.

Effect of Screw Speed. Screw speed has significant effect on both polymeric matrix and carbon particles. Increasing the screw speed resulted in more shear rate applied on the processed polymer, whereas its viscosity is decreased because polymeric melts are shear thinning fluids (pseudo plastic). Also increasing the shear rate decreases the size of the carbon particles and therefore increase its surface area and minimize the number of formed voids, these changes occurred to HDPE matrix and the carbon particles, which in turn affect directly the mechanical properties of the blend. These are shown in Figures 7–9, respectively.

TABLE 4
Effect of processing temperature on mechanical properties of 5 wt% carbon particles (≤150 µm)/HDPE blend extruded at 17 rpm

Temperature (°C)	Modulus (Mpa)	Tensile strength (Mpa)	Ductility (%EL)
200	920.0 ± 31.58	26.80 ± 1.23	11.30 ± 0.76
220	1351 ± 139.0	30.68 ± 1.50	5.30 ± 0.64
240	1295 ± 52.50	29.45 ± 0.89	5.32 ± 0.54

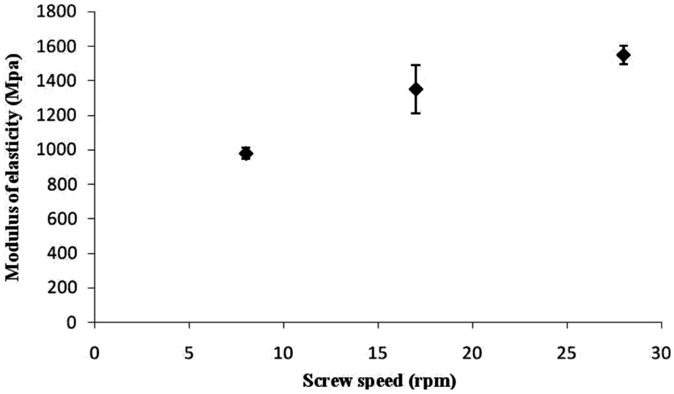

FIG. 7. Effect of screw speed on modulus of recycled HDPE/carbon particles blend at temperature value of 200°C.

It is obvious from Figures 7 and 8 that modulus and tensile strength increased by increasing the screw speed rotation. This trend is related to the decrease in polymer viscosity and increases its fluidity and ability to wet the surface of the carbon particles and enhance the adhesion between carbon particles and the polymer. This also eliminates the presence of voids by expelling of gases or air at high screw speed. Moreover as particle sizes decrease with increasing screw speed, then the interfacial adhesion and compatibility between both polymeric matrix and solid particles was improved and hence increase both the tensile strength and modulus of recycled HDPE. Also it is clear that ductility is decreased with increasing screw speed, which is a normal relation opposing an increase in modulus obtained by rising the screw speed.

Effect of Processing Temperature. The effect of processing temperature on the mechanical properties of the extruded HDPE/carbon particles blends is illustrated by Figures 10–12. It is very clear in Figures 10 and 11 that

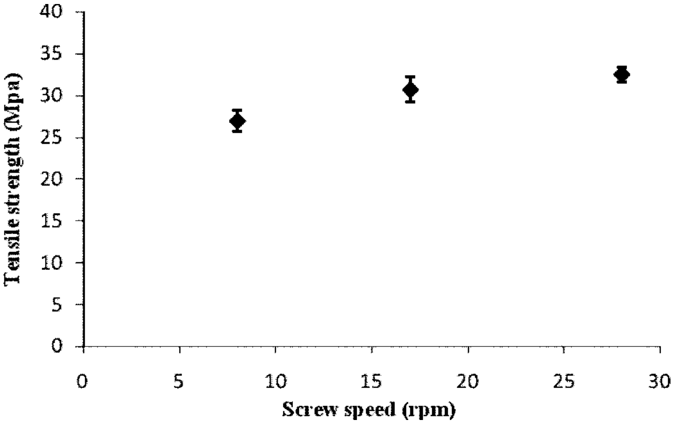

FIG. 8. Effect of screw speed on tensile strength of recycled HDPE/carbon particles blend at temperature value of 200°C.

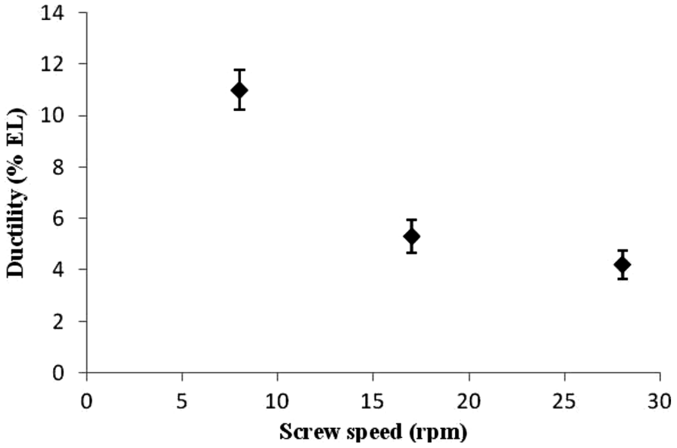

FIG. 9. Effect of screw speed on ductility of recycled HDPE/carbon particles blend at temperature value of 200°C.

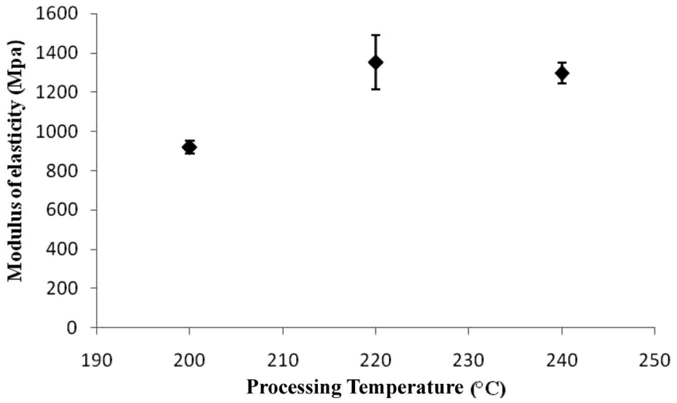

FIG. 10. Effect of processing temperature on modulus of recycled HDPE/carbon particles blend at 17 rpm screw speed.

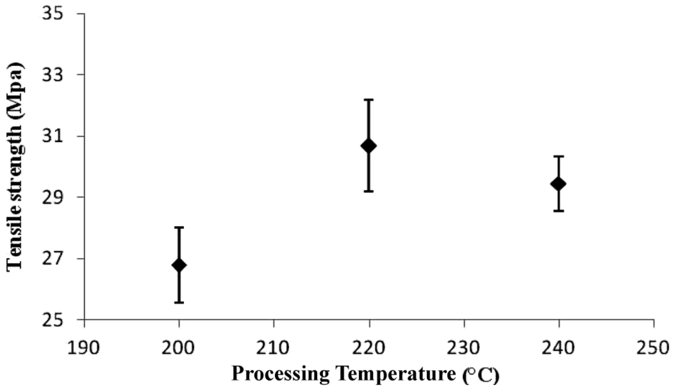

FIG. 11. Effect of processing temperature on tensile strength of recycled HDPE/carbon particles blend at 17 rpm screw speed.

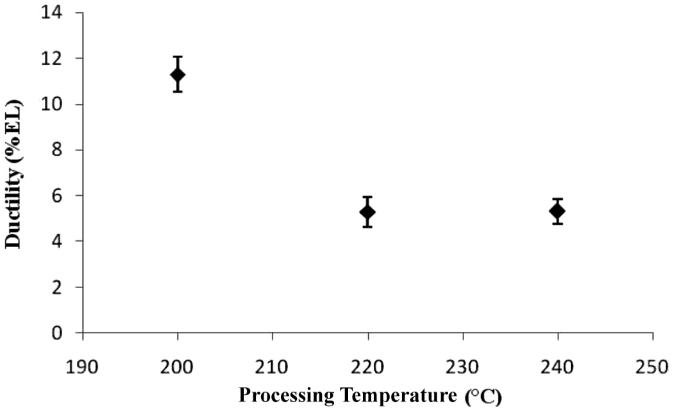

FIG. 12. Effect of processing temperature on ductility of recycled HDPE/carbon particles blend at 17 rpm screw speed.

the modulus of elasticity and tensile strength increase with processing temperature up to 220°C. This is due to the decrease in polymeric melt viscosity which has an acceptable ability to wet the carbon particles and increase their coherent with the polymeric matrix. Increasing the temperature to 240°C (a little above 220°C) a slight decrease in mechanical properties (modulus, strength) resulted due to polymeric degradation. The ductility decreases with increasing the temperature but the slope of its decrease is negligible at temperature above 220°C. This is shown in Figure 12.

CONCLUSIONS

From the work presented in this paper, the following conclusions can be drawn:

- The carbonizations of the abundant olive solid waste into carbon at 600°C in the absence of oxygen eliminates the volatile matters and increase the internal surface area of carbon, resulted in more porous carbon structure. This was carried out after several treatments to the olive solid waste. The resulted carbon of different particle sizes was used to enhance the mechanical properties of recycled HDPE. It was found that carbon particles of diameters less than 150 μm improve mechanical properties of recycled HDPE better than those of larger ones (180 μm–250 μm). Also, it was found that more carbon content can be used with the small particle size than that of large particle size. This is due to the amount and size of voids formed in the final product.
- It was found that carbon particles less than 150 μm can be added up to 5 wt% with noticed improvement in modulus of elasticity and tensile strength, while the optimum value of modulus of elasticity was found to be at 2.5% for the particles of size range of 180 to 250 μm.
- Extruder operating variables (screw speed and temperature) was found to have an effect on the mechanical properties of the recycled HDPE plastic. Tensile

modulus and tensile strength values were increased with increasing screw speed. This was related to melt viscosity and reduction in particle size with increasing screw speed. An increase in processing temperature was found to improve mechanical properties up to certain point where degradation of polymeric matrix begins to occur. Morphological studies of the surface of prepared samples by scanning electron microscope is needed to investigate the compatibility of HDPE/carbon blend.

- Finally, from the results of this work, an optimum value of carbon particles content of 5 wt% of carbon particles size of less that 150 μm was found at 28 rpm and 220°C in a single screw extruder. For particles size of the range 180–250, the optimum content is found to be 2.5% at same screw speed and temperature. All results presented in this paper are for carbonization temperature of 600°C. It is worth studying effect of carbonization temperature on the tensile properties. For industrial application, it is worth studying the electrical properties, such as insulation resistance, of the HDPE/carbon blend at various operating parameters.

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