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Improvement of Tensile Properties of Recycled Low-Density Polyethylene by Incorporation of Calcium Carbonate Particles

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ABSTRACT

Plastics are used in versatile applications including automobile, packaging, piping and house goods, these huge uses attribute in the consumption of the oil reserves and the emerged waste harm the environment when it disposed irregularly. Recycling of plastics is one of the realistic solutions to the aforementioned problems and to reduce production cost. However, the reduction in mechanical properties of recycled plastics limit their use and thus reinforced plastics become popular because of their high mechanical, physical and thermal properties. The effects of calcium carbonate content from 0 to 15 wt.% on the tensile properties of recycled low-density polyethylene (RLDPE) were tested, the addition of calcium carbonate particles up to 15 wt.% was found to enhance the tensile strength and modulus of elasticity of RLDPE samples. Three calcium carbonate particle sizes (80, 200 and 500 µm) were mixed with RLDPE to investigate the effect of particles size on the tensile properties of RLDPE, it was found that the addition of small filler particles resulted in a noticeable improvement of tensile strength and modulus of elasticity of RLDPE compared with large filler particles. It was also observed that the addition of stearic acid slightly improves tensile properties of RLDPE which may be related to improvement of the interfacial adhesion between the filler and RLDPE. The crystallization temperature and the degree of crystallinity of RLDPE were increased by the addition of 7.5 wt.% calcium carbonate particles because they act as nucleating agents.

Keywords--- Recycled Low-Density Polyethylene, Calcium Carbonate, Stearic Acid, Tensile Properties, Thermal Properties

I. INTRODCUTION

Most of the synthetic polymers cause serious environmental pollution because they are generally non-biodegradable or their biodegradation process is very slow. In order to solve this problem, scientific research was directed to recycling these polymers to produce new products that can be used in many applications. Since the properties of polymers are decreased during the recycling stage, due to the thermal degradation and/or to the applied shear stresses, so the addition of suitable reinforcements is a suitable method to improve their properties.

Different types of reinforcements were used to improve the mechanical properties of polymeric matrices such as polypropylene (PP), high density polyethylene (HDPE) and low-density polyethylene (LDPE). The mechanical properties of HDPE such as tensile strength, hardness and flexural properties were enhanced by the addition of palm fibers, but the degree of improvement was found to be affected by the fibers content, their orientation and the alkaline treatment of the fibers [1]. The effects of addition of wood flour, fine aggregates and solid olive waste on the mechanical properties of recycled polystyrene (PS) and recycled high density polyethylene were investigated. The hardness of the produced composites was improved by the addition of the reinforcements, but the degree of improvement was found to be affected by the reinforcement type and its content. [2]. Talc and carbon black were used to reinforce HDPE, the tensile strength was decreased with the increase of both types of fillers, whereas, the modulus of elasticity and hardness was increased. The flexural strength increased with the increase of carbon black content by 78% but decreased with the increase of talc content by 18% [3].

The effects of using fly ash as a filler for HDPE from economic and commercial points were investigated, the source and composition of fly ash were studied. Fly ash was a cost-effective filler for HDPE, moreover, it could improve its mechanical, thermal and electrical properties [4]. Both tensile and flexural strength properties of HDPE were found to increase with the addition of fly ash, but composites with smallest size fly ash particles proved to be better in enhancing tensile and flexural strengths [5].

The effects of modified nano-titanium dioxide on morphological, thermal, and mechanical properties of PP and HDPE were studied. Malic anhydride grafted styrene ethylene butylene styrene and silane were used as surface modification, respectively. Both tensile strength and modulus of elasticity were improved while impact strength and elongation at break were decreased. The melting and crystallization temperatures did not change very much while the degree of crystallization was increased about 25-30% [6].

The role of stearic acid content in crystallization and thermal behavior of HDPE/calcium carbonate composites was studied. Crystallization and melting temperatures of HDPE with addition of 10 wt.% calcium carbonate were increased, while the addition of stearic acid decreased crystallization temperature and had no significant effect on melting temperature [7].

In this study; calcium carbonate particles were used to improve the tensile properties of recycled low-density polyethylene (RLDPE) which causes harms to environment especially when thrown into lands or burned irregularly. The effects of the calcium carbonate particles content, their size and the addition of stearic acid on tensile and thermal properties of recycled LDPE were investigated.

II. MATERIALS AND METHODS

2.1 Materials

LDPE and calcium carbonate were supplied from Royal Industrial Plastic Company and Palestine Plastic Industries Company, respectively, whereas stearic acid with purity of 95% was purchased from Sigma Aldrich. Calcium carbonate was sieved, three grades of particle sizes selected were with diameters of 80, 200, and 500 µm, the selected particles were then oven-dried under vacuum for 4 hours at 70 ℃ to reduce their moisture content.

2.2 Samples Preparation

The samples were produced by using a homemade two-roll machine, it consists of two stainless steel rolls that rotate in opposite directions at a fixed speed of rotation, it is equipped with electrical heaters in order to melt the polymeric materials, the processing variables such as the processing temperature, the speed of rotation, the distance between the rolls and the mixing time can be controlled. At the first stage, virgin low-density polyethylene (VLDPE) pellets were melted using a two-roll mill machine at 140°C at low speed for 5 minutes, then it was cooled to room temperature. In the second stage, the samples obtained from the first stage was melted at the same processing conditions in order to investigate the effect of recycling process on its mechanical properties. Finally, RLDPE was melted by using a two- roll mill machine at 140°C at low speed for 3 minutes and then calcium carbonate particles with or without stearic acid and mixed for 2 minutes.

A homemade thermal press molding apparatus 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. Each sample was pressed into 2.0 mm thick sheets using 8×10 cm mold at 140 °C for 10 minutes under a pressure of 5-6 bar. After that, the produced sheet was cooled to room temperature by using a cooling water system.

2.3 Samples Characterization

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/min. For each composite material, five specimens of 70 mm gauge length, 42 mm width and 2.0 mm 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

Thermal properties of the produced samples were measured by using differential scanning calorimeter (DSC) model Pyrix-6, PerkinElmer Corporation, U.K. A mass of 5 mg from each sample was placed in a sealed aluminum pan, the same temperature profile (from room temperature to 200 ^{0}C), heated at 10°C/min 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. The latter was 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).

III. RESULTS AND DISCUSSION

3.1 Tensile Properties

Table (1) shows the tensile strength and modulus of elasticity of virgin and recycled LDPE both properties were decreased due to recycling action, the decrease in the tensile properties may be related to thermal degradation of the polymer. Calcium carbonate particles having a diameter of 50 µm was mixed with RLDPE in order to improve its tensile properties. Figures 1 and 2 show the effect of calcium carbonate content on the tensile strength and modulus of elasticity of RLDPE/calcium carbonate-based composites.

Table 1: Tensile strength and modulus of elasticity of VLDPE and RLDPE

Sample	Tensile strength (MPa)	Modulus of elasticity (MPa)
VLDPE	7.28±0.61	229.1±34.47
RLDPE	6.68±0.05	111.00±6.71

It can be observed from Figure 1 that the tensile strength of RLDPE/calcium carbonate-based composites decreased with increasing the content of calcium carbonate particles to reach its minimum value at 7.5wt.%, then it starts to increase to reach 9.09 MPa when 15.0wt.% of calcium carbonate particles were added. The initial decrease in the tensile strength may be related to the poor mixing and distribution of the filler particles which may act as stress concentrated points. On the other hand, the addition of calcium carbonate particles enhanced the modulus of elasticity of RLDPE, it can be seen from Figure 2 that the addition of calcium carbonate particles increased the modulus of elasticity from 111.00 MPa to 273.57 MPa. 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 may be related to fact that the filler acts as load carrier. On other hand, an increase in degree of crystallinity of RLDPE was achieved as shown in Table 4.

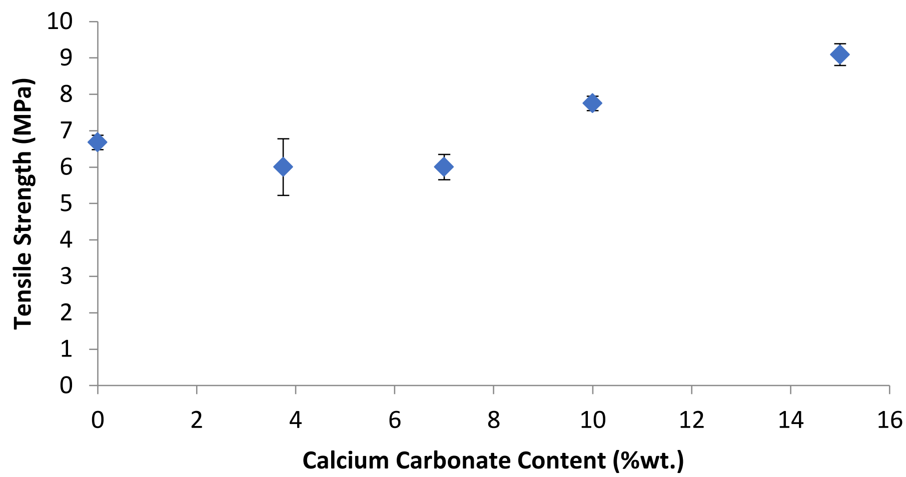

Figure 1: Effect of calcium carbonate content on the tensile strength of RLDPE-calcium carbonate-based composites

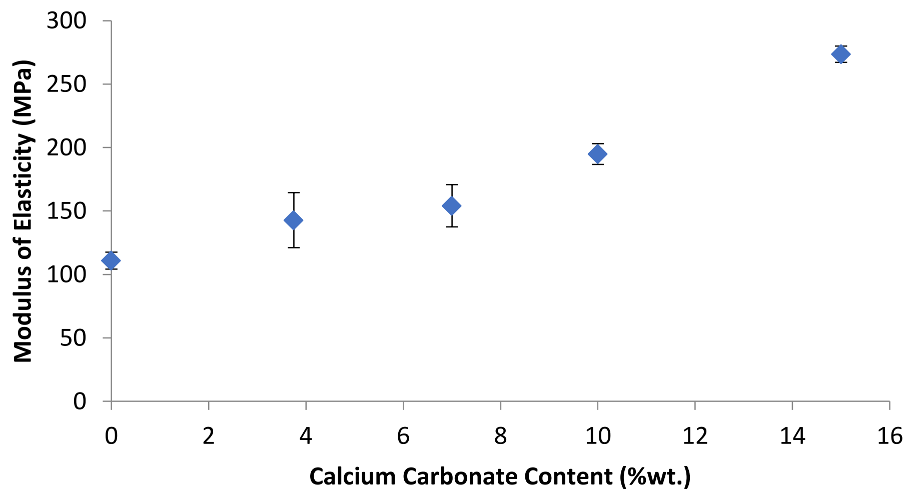

Figure 2: Effect of calcium carbonate content on the modulus of elasticity of RLDPE-calcium carbonate-based composites

It is clear from the results that the addition of 15 wt.% of calcium carbonate particles improves the tensile properties of RLDPE, and the produced composites have better tensile properties than VLDPE. Organic and synthetic reinforcements such as palm fibers, glass fibers and fly ash were found to improve the mechanical properties of HDPE [1, 4].

In order to investigate the effect of calcium carbonate particles size on the tensile properties of RLDPE, three samples containing different particles size were produced at a constant calcium carbonate content (7.5wt.%).

Table 2: Effect of calcium carbonate particles size on the tensile strength and modulus of elasticity of RLDPE

Calcium carbonate particles diameter (µm)	Tensile Strength (MPa)	Tensile Modulus (MPa)
-----	6.68±0.05	111.00±6.71
80	8.63±0.10	252.04±12.01
200	6.48±0.55	181.64±21.09
500	6.00±0.35	154.12±16.58

It can be observed from Table (2) that the tensile strength and modulus of elasticity of RLDPE are inversely proportional with the particles size. Calcium carbonate particles having a diameter of 80 µm have better results and can improve both the elastic modulus and the tensile strength up to values of 252.04 and 8.63 MPa, respectively. The addition of 7.5wt.% of calcium carbonate particles increases the tensile properties of the RLDPE with decreasing the particles size due to the large surface area of these particles which positively 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 [1, 5, 8].

The interfacial adhesion between the reinforcement and the polymeric matrix affects strongly the mechanical properties of the composite materials because the applied stress transfers from the matric to the reinforcement via the interface. In order to improve the interfacial adhesion between RLDPE and calcium carbonate particles, stearic acid was added at a constant calcium carbonate content (7.5 wt.%) and a constant particles size (80 µm). It can be observed from Table (3) that the addition of stearic acid improves the tensile properties of the produced composite materials which may be related to the improvement in the interfacial adhesion between filler and the polymeric matrix. The improvement in the tensile strength and modulus of elasticity of composite materials by using suitable coupling agents was reported in previous studies [6,9].

Table 3: Effect of stearic acid content on the tensile strength and modulus of elasticity of RLDPE

Stearic acid content (wt.%)	Tensile Strength (MPa)	Tensile Modulus (MPa)
0.0	6.00±0.35	154.12±16.58
7.0	6.87±0.25	292.6±11.37
10.0	7.04±0.65	208.42±15.56

3.2 Thermal Properties

Table (4) shows the melting point, the crystallization temperature, the heat of fusion and the percentage of crystallinity of VLDPE, RLDPE and RLDPE containing 7.5wt.% calcium carbonate particles having 80 µm diameter (RLDPE-CC). The percentage of crystalline regions where the last can be calculated from the following equation:

$$x_c = \frac{\Delta H_f}{\Delta H_f^0 X \varphi} X100\%$$

Where X_C is percent 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 [10] and φ is weight percent of polyethylene in the produced composite.

Table 4: Thermal properties of VLDPE, RLDPE and RLDPE containing calcium carbonate particles (RLDPE-CC)

Sample	Melting Temperature (^{0}C)	Recrystallization Temperature (^{0}C)	ΔH^0_{fusion} (J/g)	%Crystallinity
VLDPE	108.08	88.56	71.05	24.5
RLDPE	107.17	89.97	68.66	23.7
RLDPE-CC	106.70	95.16	88.28	32.9

It can be observed from Table (4) that the melting temperature and degree of crystallinity of RLDPE are lower than that for VLDPE which may be related to the degradation of the polymer due to the processing temperature and the applied shear stresses during the recycling process. The addition of calcium carbonate particles improves the recrystallization temperature and degree of crystallinity of RLDPE which may be related to the fact that calcium carbonate particles act as nucleating agents. It is worthy here to point that other researchers had found that the degree of crystallinity of both PP and HDPE was found to increase by the addition of titanium dioxide and silicone dioxide-calcium oxide-magnesium oxide, respectively [6-9]. The melting point of RLDPE was slightly decreased by the addition of 7.5 wt.% calcium carbonate particles which in its turn will positively affect the ease of processing of these composite materials.

IV. CONCLUSION

It can be concluded that the addition of calcium carbonate particles enhanced both the tensile strength and the modulus of elasticity of RLDPE, the degree of improvement was found to be dependent on the calcium carbonate particles content and their size. The maximum values were obtained when 15 wt.% calcium carbonate particles were used, while the addition of particles having 80 µm diameter resulted in better tensile properties compared with other particle sizes. The addition of stearic acid was also found to enhance the tensile properties of RLDPE. On the other hand, the crystallization temperature and the degree of crystallinity of RLDPE were improved by the addition of 7.5 wt.% calcium carbonate particles.

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