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Noora Al-Qahtani (✉ noora.alqahtani@qu.edu.qa)

Qatar University

Maryam Al-Ejji

Qatar University

Mabrouk Ouederni

Qatar Petrochemical Company

Mariam AlMaadeed

Qatar University

Nabil Madi

Qatar University

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Effect Of Carbon Black Loading On Linear Low-Density Polyethylene Properties

Noora Al-Qahtani^{1,*}, Maryam Al-Ejji¹, Mabrouk Ouederni², M. A. AlMaadeed¹, and Nabil Madi¹

¹ Center for Advanced Materials, Qatar University, Doha-Qatar, P.O. Box: 2713.

² Resercher and Development of Qatar Petrochemical Company, QAPCO, Doha-Qatar P.O. Box 756.

Key words: Carbon Black, Electrical properties, Thermal analysis, Reheological properties.

***Correspondence to:** Dr. Noora Alqahtani (noora.alqahtani@qu.edu.qa)

ABSTRACT

The mechanical, thermal, electrical, rheology, and morphological properties of composites made from linear low-density polyethylene and carbon black (CB) content of 5%, 10%, and 20% by weight were investigated. The optimum mechanical and electrical properties performance was achieved with the adding of 5% carbon black. The drop in properties after adding more CB is due to agglomeration and poor dispersion of carbon particles in the polymer matrix. CB resulted in higher dynamic viscosity and storage modulus at low frequencies, although this phenomenon was reversed at high frequencies. CB resulted in more shear thinning of LLDPE at high shear rates.

1. INTRODUCTION

Linear low-density polyethylene is commonly utilized polymer in the industry because of its unique structure and excellent overall performance. It is produced by polymerizing ethylene with a slight amount of an α -olefin. LLDPE, however, is not photodegradation resistant and its relatively low mechanical properties and thermal stability can sometimes limit its application in industry. The development of particulate reinforced polymer composites is currently viewed as one of the highly promising methodologies in the area of next generation engineering products [1–3].

Polymer composites are an essential category of polymers that has broad applications in a number of various industrial areas and therefore have been comprehensively researched in the last few decades [4–7]. Inorganic fillers are believed to be extremely critical both technologically and economically, including carbon black which is very valuable in the thermoplastic industry [8]. Further the development of electrical conductivity in low carbon black content [9], the enhancement of mechanical and thermal properties is of particular concern. The high feature percentage in mixture with high particle porosity, high strength, and compatibility with organic materials constituted carbon black a potential candidate as support for polymeric materials. An effective development of the carbon black properties in polymers is consequently associated to their homogeneous dispersion in the matrix. The carbon black dispersion is strongly dependent on carbon black content and therefore a percolation threshold value for the composite should be investigated. Carbon black used as fillers in many polymers such as ultra-high molecular weight polyethylene (UHMWPE) [10,11], high density polyethylene (HDPE) [12,13], ethylene butylacrylate copolymer (EBA) [14], ethylene–octene elastomer (EO) [15], low molecular weight polyethylene (LMWPE) [16], Low-density polyethylene (LDPE) [17] and Polypropylene (PP) [17]. The fillers were around of 0.5wt% to 5wt%.

The addition of CB to LLDPE has been investigated by many authors due to its low cost and conductive properties [18–20] [18-21]. Pin Zhou et al. [18] experiment the electrical properties of the CB filled LLDPE/EMA composites and higher percolation threshold due to the good interaction between EMA and CB. The stability of CB filled LLPE is important and studied by V.M Goldberg et al. [18] for thermo oxidative degree and artificial weathering by M.liu et al. [19] both showed that CB has a positive impact on the stability. Mechanical properties of CB filled LLDPE was studied by A.Ahmed et al. [21] and showed the electron irradiation important the properties at 250 kGy. This paper discuss the impact of CB on the mechanical, electrical, thermal stability and rheology of LLDPE. The relation between the microstructure and dispersion of CB is explained.

2. EXPERIMENTAL

2.1. Materials

The materials utilized in this research were Linear Low-Density Polyethylene (LLDPE) provided as pellets by Qatar Petrochemical Company (QAPCO) with the following properties, a Melt Flow Index (MFI) of 1.0 g/10 min (190 °C, 2.16 kg) and a density of 0.918 g/cm³. The average particle

size of CB was about 30 nm and a density of 2.5 g/cm³. The Materials were completely dried before the combination that reduce the impact of moisture [22].

2.2. Composite sample preparation

Composite samples were arranged in a two- step process that included compounding and molding. First LLDPE and CB powders were tumbled mixed in the dry state in the right proportions before being compounded in a Laboratory size FARREL 2 Roll Mill. The mill temperature was set at 160 °C and samples were fed twice into the Mill for a total compounding time of about 12 min. The second step consisted of molding 2 mm thick composite sheets in a Compression Molding Press Machine. The molding cycle consisted of the following: 5 min at a temperature of 180 °C and zero pressure, followed by 5 min at T = 180 °C and P = 1400 psi, and finally about 5 min cooling at a rate of about 15 °C/min. Table 1 indicates the defined symbol for each method and the subsequent compositional ratio of each component.

Table 1: Samples code and their compositions

Designated Symbol	Composition and compositional Ratio (By Weight %)
LLDPE	100 PE/0 CB
PE5CB*	95 PE/5 CB
PE10CB	90 PE/10 CB
PE20CB	80 PE/20 CB

*Abbreviations consist of PE and CB stands for LLDPE and carbon black respectively.

2.3. Differential Scanning Calorimeter (DSC) Measurements

Differential scanning calorimeter assessment was carried out applying a Perkin Elmer Instrument Pyris 6 (DSC) with a sample weight of 5 to 8 mg. All samples were heated at a rate of 10 °C/min to 170 °C, next held for 5 min at 170 °C to remove the earlier thermal history, and then cooled at a rate of 10 °C/min to 30 °C, then held for 5 min at 30 °C and heated again at a rate of 10 °C/min to 170 °C under nitrogen atmosphere (20 ml/min) [22]. The cold crystallization temperature (T_c °C), melting temperature (T_m °C), and heat of fusion were revealed from the second heat scan. The crystallinity of samples (X_c %) was determined employing the subsequent expression [23]:

$$X_c \% = \frac{\Delta H}{\Delta H_f W} \times 100 \quad (1)$$

The crystallinity was determined from the ratio among the melting enthalpy (ΔH) amount from the second scan of the DSC assessment and the melting enthalpy of the 100% crystalline phase (ΔH_f) from the literature [24]. For polyethylene a ΔH_f value of 289 J/g was utilized [24], and W is the weight fraction of LLDPE in the composites. Half crystallization time ($t_{1/2}$) is the time necessary to 50% of crystallization to be done, which represent the speed of crystallization rate [25], $t_{1/2}$ can be obtained by the following equation [26]:

$$t_{1/2} = \frac{(T_{c.onst} - T_c)}{r} \quad (2)$$

Where, ($T_{c.onst}$) is the onset temperature of crystallization process, (T_c) is the crystallization temperature, and (r) is the cooling rate (r).

2.4. Thermogravimetric Analysis (TGA) Measurements

Thermogravimetric (TG) and Differential Thermogravimetric (DTG) measurements were performed with Perkin Elmer Pyris Analyzer 6 [22]. The specimens of LLDPE and composites with weight of about 10 mg were heated from 30 °C to 550 °C at a heating rate of 10 °C/min [27]. Samples were arranged in a ceramic pan and the tests were done in a nitrogen atmosphere with N_2 being provided at a flow rate of 20 ml/min. In this work, comprehensive and precise aspects identifying the thermal stability based on the temperature at which sample losses 50% of its weight ($T_{50\%}$) and the temperature of maximum rate of weight loss (T_{peak}) (from DTG curves) are carried as a measure of thermal stability of the composites [28].

2.5. Tensile test Measurements

The tensile properties of the composites were calculated as per the standard test method ASTM D 638-02 [29]. The tensile strength of the composite samples were tested by Lloyd instrument's material analysis machine related to a remote microcomputer for data acquisition and evaluation. The load was assessed by a load cell of 50 kN capacity, although the displacement was evaluated applying an internal extensometer. The speed of examination was 100 mm/min [30]. Samples were examined under the similar requirement for each category of samples. The values described were the average values of three individual amounts [31].

2.6. Hardness Test Measurements

A micro - hardness analysis was organized with all samples by HRF - Rockwell Hardness F Scale by application of load 60 kg and the provided measurement values were of indentation hardness of a total of 10 indentation points on the sample surface [32]. The slanted was utilized to calculate the hardness of the compounds corresponding to ASTM D785-08.

2.7. Morphological Observations

The microstructures of composite samples were examined applying a scanning electron microscope (Nova Nano SEM 450) [33] . These samples were fractured in liquid nitrogen into pieces with a surface area of 2mm, each surface of which was coated with a thin layer of gold and then positioned into the SEM. The microstructure of the samples was examined at an accelerating voltage of 3kV. Through the microstructural examination, the interactions among CB and virgin polymer were found [34].

2.8. Rheology Characterization

The viscoelastic behavior of the neat polymer and CB reinforced samples was analyzed by melt rheology in an AERS Rheometer. Samples were first molded into 2 mm thick disks and then rheology measurements were presented to define the storage modulus (G'), loss modulus (G'') and dynamic viscosity (η^*). Experiments were finalized over a frequency sweep range among 0.01 and 100 rad/s at a temperature of 210 °C [35] .

2.9. Electrical Conductivity Measurement

The electrical conductivity was assessed by Keithley 2400 source meter using the two-probe method. The electrical measurement was carried out at room temperature 25 °C. The samples were formulated as disks of 2 mm thickness and covered with silver sticker on both sides of the disc to ensure good contact of the sample surface with the two aluminum electrodes (1 mm diameter) which fixed on the opposite sides of the sample [36]. Figure 1 illustrates the measurement scheme. The specific conductivity σ was measured by means of a device measuring very high resistance. The procedure was set different values of current for measuring the voltage and calculating the resistance (R) [10]. Equation (3) is used to calculate the conductivity where A is the area of discs samples and L is length of disk.

$$\frac{1}{\sigma} = R \frac{A}{L} \quad (3)$$

Figure 1: Illustration of the measurement for the electrical conductivity.

3. RESULTS AND DISCUSSION

3.1 Differential Scanning Calorimeter (DSC) Measurements

The heating and cooling thermograms of the LLDPE/CB composites are shown in Figure 2. The DSC findings in terms of melting point (T_m , °C), crystallization temperature (T_c , °C) and the percentage crystallinity (X_c) are summarized in Table 2. The DSC measurements indicate that the melting point of composites is not considerably influenced by variations in the CB substance. Furthermore, the X_c of composites is not affected by a CB addition of 5 wt% but then decreased with 10 and 20% of CB addition. A low shoulder might be observed in the low point of temperature side of the melting bend of the LLDPE indicating paracrystalline structure. A virgin LLDPE has a relatively wide molecular weight distribution and small series branching arrangement. The branches are rather found in the lower molecular weight chains; hence, LLDPE performs as is a combination of higher molecular weight linear and lower molecular weight branched particles [37]. This heterogeneous distribution of polymer chains is considered as a wide melting zone. The CB fragments are recognized to nucleate the polymer crystallization, growing its crystallinity and affecting the nature of the lamella in the crystallite [38]. As can be seen, from Figure 2 and Table 2 this nucleating function of the carbon particles in the crystallization of LLDPE is small. At a CB particle of extra than 5 wt% the reduction to crystallinity level can be justified by the existence of an extreme amount of CB contents that can obstruct the movement of the polymer chain parts and therefore, prevent crystal growing [28,20]. This can be explained by the fact that crystallization is composed of both nucleation and growth. Even though nucleation is enhanced by the CB particles, growth is significantly reduced due to restrictions on mobility of the polymer chains in the occurrence of a high concentration of CB

particles. The half crystallization time $t_{1/2}$ was enhanced with the addendum of CB content and is generally in agreement with the crystallinity results discussed earlier. It can be concluded that the nucleation started and the crystallization procedure progressed in a relatively short period for the neat polymer [25]. The smaller time of $t_{1/2}$ represent the faster crystallization rate [39]. The slow crystallization rate of CB composites can be illustrated by the weak CB-polymer interactions [40] and slow crystal growth in the presence of CB particles.

Table 2: Melting Temperature, Crystallization Temperature, % Crystallinity and half crystallization time of CB composites.

Sample ID.	T _m (°C)	T _c (°C)	X _c (%)	t _½ (min)
LLDPE	122.3	106.1	42.1	0.26
PE5CB	122.3	106.3	42.1	0.3
PE10CB	121.7	106.7	31.1	0.27
PE20CB	122.2	106.3	30.3	0.29

Figure 2: DSC thermo-gram of LLDPE and its composites with carbon black for -melting and cold crystallization profiles.

3.2 Thermogravimetric Analysis (TGA) Measurements

DTGA curves of pure LLDPE and LLDPE composites with various CB loadings are exhibited in Figure 3. It is viewed that the thermal decomposition of all mixtures happens below 500 °C. The temperature at which a sample drops 50% of its weight ($T_{50\%}$), these were taken from TG curves (not shown), and temperature of max rate of weight loss (T_{end}) are carried as an amount of thermal stability of the composites and represented in Table 3. It is apparent from Table 3 that, adding CB improves the thermal stability of the composites. The noted growth in thermal stability is believed to be attributable to the limitation of mobility of segmental progress of LLDPE, because of the improved interface among the CB and the polymer matrix [40–43].

Table 3: Change of TGA profiles with different contents in the LLDPE composites

Sample ID.	Temperature (°C)	
	$T_{50\%}$	T_{end}
LLDPE	470.1	475.7
PE5CB	472.6	477.4
PE10CB	475.0	479.1
PE20CB	476.3	481.0

Figure 3: Change of DTGA profiles with varying contents of CB.

3.3 Tensile test Measurements

Figure 4 indicates the difference in the tensile strength with the addition of CB. The tensile strength increases significantly by adding CB at 5 wt% and then it decreases rapidly when CB content is more than 5 wt%. This indicates that the optimum carbon black loading in LLDPE is about 5 wt%. The degree of reinforcement of LLDPE composites increases with filler loading and the extent of polymer-filler interaction. The reduction of tensile strength beyond 5 wt% loading can be explained in terms of particle agglomeration at higher content of CB [44,45]. These agglomerated, hard particles, create concentrated stress points and therefore reduce the stress transfer from polymer chains to carbon particles [20]. So, at higher content, CB particles will exhibit a poor dispersion and it is evidently difficult to shape strong adhesion, weak surface energy, with polymer matrix. This will be shown later in the morphological study.

The influence of CB loading on the samples modulus is shown in Figure 5. The neat polymer, LLDPE, is of low tensile modulus 141 MPa, after adding CB the tensile modulus increases with increasing CB contents, which reveals enhanced stiffness caused by increasing filler substance. A change of distortion performance from ductile to more brittle occurred at higher fillings of CB. As the amount of CB particles increases in the LLDPE matrix, some portions of the polymer are trapped inside the filler network, which increases the effective volume ratio of the solid particles in the composite. On the other hand, polymer-filler interface might cause in the absorption of the polymer chains on the filler surface and reduces its progress. Consequently, the polymer viscosity and modulus will be increased [46,47].

From the point of view of packaging technology, agricultural film applications and extension at break is a critical factor explaining the ductility of materials below tension. Figure 5 reveals the need of elongation at break of the mixtures on the CB filling. CB loading of 5 wt% resulted in an elongation at break of PE5CB composite of 1200 %, subsequently its quick reduction with rising CB content. The results indicate that the CB may possibly substantially develop the tensile fracture ductility of LLDPE resin, and the best effect was obtained when CB was 5 wt%. The breakdown of the filled combinations with polymer matrix can be introduced through minor defects initiated by stress intensity on the filler surface. Therefore, little cracks are founded which expand till achieving the vital crack amount. In the identical material without the filler, the creation and expansion of cracks are unintentional, and the collapse appears at reasonably high distortion. The existence of the filler findings in a simpler start of break formation by a stress intensity on the filler surface, on one hand and on the other hand a reduce of chain

mobility as a result of the polymer-filler interface causing in a lower deformability of the combinations. Both consequences cause to a reduction of the elongation at break amounts [45,48].

Figure 4: Tensile Strength of CB reinforced LLDPE composites.

Figure 5: Young's Modulus and Percentage Total Elongation at Fracture for the composites.

3.4 Hardness Test Measurements

Surface hardness is usually examined as one of the extremely vital elements associated to the wear resistance of materials. Figure 6 shows the hardness as a function of CB content in LLDPE. It shows that the hardness of the composite is almost linearly enhanced with rising CB content. This indicates that addition of CB particles is beneficial for improving surface hardness of LLDPE composites. CB particles acted as stress concentration points. It is expected; therefore, beyond 5 wt% of CB content, polymers chains have penetrated into the CB particles agglomerates [49].

Figure 6: Hardness for LLDPE composites.

3.5 Morphological Observations

Figure 7A shows the SEM micrographs of the CB by itself. As shown in the figure, the average size was around 30 nm. Figures 7B-E show the freeze fractured morphology of pure LLDPE and 5,10 and 20 wt% CB filled LLDPE respectively. Figures 7C-E show SEM micrographs of agglomerates typically found in these composites. Evidently as shown in Figure 8 C, 5 wt% loading, CB particles are distributed in the polymer matrix. As a result, the tensile strength of LLDPE is significantly improved. The used LLDPE has a relatively low melt flow index (MFI = 1.0 g/10 min), and a flow behavior that favors in good dispersion of CB in the polymer matrix especially at low CB polymer ratio (lower loading). However, as the CB content rises, CB atoms blend into main aggregates, and Van der Waals forces affect these aggregates to enter in additional loosely assembled agglomerates [20,50], as might be viewed from Figures 7D and E. So, at higher filler loading one expects a reduction in the tensile strength (c.f. Figures. 4)

(A)

(B)

(C)

(D)

(E)

Figure 7: SEM micrographs of the fracture surface of the specimens with various filler contents
(A) CB structure, (B) Virgin LLDPE, (C) CB 5wt%, (D) CB 10wt%, (E) CB 20wt%.

3.6 Rheology Characterization

The rheology characterization of the various composite samples gives a very good indication of their behavior in an extrusion or a molding process. The rheology also provides further understanding of the effect of CB on the LLDPE composites. As we can see in Figure 8, the flow behavior of LLDPE is significantly affected by the addition of CB. This is true both at low and high frequencies. The storage modulus of the LLDPE melts (G') increases by about 50 times just by the addition of 5% CB, in the case of 20% CB it jumps by 130 times. This elastic behavior of the melt is reversed at high frequencies and the 20% CB composite shows the lowest G' at the highest frequency. This again supports the earlier observation regarding the agglomeration of the CB aggregates at the high loading of 20%. The 5% composite is the closest in behavior to the virgin polymer in terms of its melt processing due to the good dispersion of the CB particles in the composite matrix. Figure 9 shows the changes in dynamic viscosity as a function of increasing frequency for the different composites. Even though the viscosity of LLDPE is increased by the addition of CB at low shear, this is not the case at high shear rates where the neat polymer maintains a higher viscosity than the composites. CB actually increases the shear thinning of LLDPE as the frequency goes up as it is seen on the right-hand side of Figure 9. Shear thinning is a known phenomenon in polyethylene [51]. The 5% addition shows the best uniformity and distribution morphology when it comes to the melt processing of LLDPE composites, and the higher loadings of CB significantly change the processability of the polymer.

Figure 8: Storage Modulus (G') as a function of frequency for a) virgin LLDPE, b) PE5CB, c) PE10CB and d) PE20CB.

Figure 9: Dynamic Viscosity (η^*) as a function of frequency for a) virgin LLDPE, b) PE5CB, c) PE10CB and d) PE20CB.

2.10. Electrical Conductivity Measurement

To investigate the effects of the filler and LLDPE content on the conductivity. Figure 10 indicates the reliance of electrical resistivity at room temperature, as a role of the comparative carbon black-amount. As observed, a greater composition of the filled CB particles findings in a lower electrical conductivity. The level of diffusion of CB in the 5CB matrix is higher than in the 10CB and 20CB. Here particular, the variation in crystallinity of the polymers performs a main part in clarifying the resistivity of the CB/thermoplastic polymer compounds explored [52]. The 20 CB have greater resistivity because of its smaller crystallinity. The rising in the filler loading more typically has slightly influence on the combined electrical resistivity. The conductivity of the mixture falls significantly with rising quantity of conducting filler in the polymer format, and the combination felt an insulator-to-conductor transition at a specific vital filler substance. This quick break down, indicating the accumulation of conducting atoms to shape systems, is well-defined as the percolation transition [13]. In an exciting research by Du and coworkers [53], they discovered that the level of alignment necessary to achieve highest electrical conductivity in the path of nanofiller alignment declines with rise in nanofiller amounts.

Figure 10: Influence of CB content on conductivity.

4. CONCLUSION

In this research, composites produced by mixing varying contents of CB nanoparticles and LLDPE were investigated. The results show that:

- a- These findings can be very useful in the design and operation of LLDPE industrial compounding technology.
- b- Thermal analysis of the composites indicated a decrease in the quantity of crystalline phase upon addition of CB. The decrease is significant for PE10CB and PE20CB samples, indicative of a lower quantity of the ordered phase with increasing filler content due to agglomeration and limited mobility in the melt during crystallization. The thermal stability is slightly improved for CB filled LLDPE composites.
- c- 5 CB is considered good conductor, increasing the fillers do not increase the conductivity.
- d- The addition of CB to thermoplastics causes a large increase in viscosity, especially at low shear rate, where at sufficiently high additives loadings, the viscosity may become unbounded due to strong interaction between the CB particles. It was demonstrated that CB increases the shear thinning behavior of LLDPE.
- e- The overall analysis of thermal, mechanical, and rheology results, leads us to recommend that the optimal loading of CB in the LLDPE composite should be around 5%.

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Figures

Figure 1

Illustration of the measurement for the electrical conductivity.

Figure 2

DSC thermo-gram of LLDPE and its composites with carbon black for -melting and cold crystallization profiles.

Figure 3

Change of DTGA profiles with varying contents of CB.

Figure 4

Tensile Strength of CB reinforced LLDPE composites.

Figure 5

Young's Modulus and Percentage Total Elongation at Fracture for the composites.

Figure 6

Hardness for LLDPE composites.

Figure 7

SEM micrographs of the fracture surface of the specimens with various filler contents (A) CB structure, (B) Virgin LLDPE, (C) CB 5wt%, (D) CB 10wt%, (E) CB 20wt%.

Figure 8

Storage Modulus (G') as a function of frequency for a) virgin LLDPE, b) PE5CB, c) PE10CB and d) PE20CB.

Figure 9

Dynamic Viscosity (η^*) as a function of frequency for a) virgin LLDPE, b) PE5CB, c) PE10CB and d) PE20CB.

Figure 10

Influence of CB content on conductivity.