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# Effects of Blend Composition on the UV Stability of Polyethylene Blends

Anour Shebani*, Abdalah klash, Rabia Elhabishi, Shalh Abdsalam, Wael Elhrari
Libyan Polymer Research Center, Tripoli-Libya
*anour@prc.ly

## Abstract

The main goal of this study was to investigate the effects of blending composition on the UV stability of different polyethylenes (PEs). This would help in having a good understanding of modifying these materials in different ways, to make them suitable for different target applications. The susceptibility of different blends of low density polyethylene (LDPE) and high density polyethylene (HDPE) to UV degradation were investigated. LDPE/HDPE blends with varying ratios (80/20, 60/40, 40/60, and 20/80), were prepared by melt-mixing technique using mini-twin-extruder. The LDPE, HDPE and their blends were conducted and exposed to UV-B light (340nm) for 415h in accelerated weathering test machine. To follow the degradation and to characterize the surface changes of LDPE, HDPE and their blends, fourier transform infrared spectroscopy (FTIR), contact angle measurements and optical microscope were utilized. It was observed that the susceptibility of LDPE/HDPE blends to degradation varied according to blending composition. Blends-rich with HDPE possess better resistance to UV degradation than virgin LDPE and blends-rich with LDPE. Moreover, blend with the composition of LDPE (20)/HDPE (80) was less susceptible to UV degradation than other blends. In line with previous study, blending composition could be used as indicator to predicate the final properties and suitable application of the final LDPE/HDPE blend.

Key words: LDPE; HDPE; UV degradation; blending; properties

## المستخص

تهدف الدراسة الى التحقق من تأثير خلط أنواع مختلفة من متعدد الإيثيلين على ثبايته عند تعرضه للأشعة الفوق بنفسجية (UV). هذا من شأنه أن يعطي نتائج تساعد في فهم تأثير عملية الخلط لتحسين خواص هذه البوليمرات لجعلها مناسبة لبعض لتطبيقات. تم في هذه الدراسة خلط نسب مختلفة من متعدد إيثيلين منخفض الكثافة (LDPE) ومتعدد إيثيلين عالي الكثافة (HDPE) ودراسة تأثير أشعة UV عليها. حيث تم تحضير خليط من LDPE/HDPE بنسب مختلفة (20/80، 40/60، 60/40، و 80/20) بواسطة ألة البثق ثنائية المحور. تم تعريض البوليمرات الاساسية والمخالط البوليمرية لاشعة UV (UV-B 340 nm) لمدة 415 ساعة باستخدام صندوق التجوية الاصطناعية. متابعة التغيرات في الخواص السطحية للبوليمرات والمخاليط المعرضة للأشعة تم بواسطة اجهزة مثل جهاز مطيافية الأشعة تحت الحمراء وجهاز قياس زاوية التلامس والمجهر الضوئي.

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أشارت النتائج إلى أن التغيرات في الخواص السطحية للمخاليط تتفاوت وفقاً لمكونات كل مخلوط بوليمري. فالمخاليط البوليمرية الغنية بـ HDPE تتميز بمقاومة أفضل لأشعة UV من LDPE والمخاليط الغنية بـ LDPE. كما بينت الدراسة بأن أفضل ثباتية وأكثر مقاومة كانت للمخلوط المكون من (LDPE (20) / HDPE (80)) مقارنة بباقي المخاليط البوليمرية. وبالتوافق مع دراستنا السابقة يتضح بأن نسب وعمليات خلط الانواع المختلفة من متعدد الإيثيلين من شأنها أن تستخدم كمؤشر لتوقع الخصائص النهائية والتطبيق المناسب للمخاليط البوليمرية (LDPE/HDPE).

## Introduction

Polyethylene (PE) is one of the most used polymer in modern society. PE is a name given to different types of polymer; low density polyethylene (LDPE), linear low density polyethylene (LLDPE) and high density polyethylene (HDPE). These grades are divided based on density and branching. They have the same chemical formulas and their overall properties depend significantly on variables such as the extent and type of branching, the crystal structure and the molecular weight [1-2]. They have been used in many applications owing to the excellent mechanical properties such as high drawability, toughness, workability, lightweight and so forth [3]. PE is a consumer polymer which can be used in applications ranging for films, tubes, pipes, plastic parts, laminates, etc. in several markets (packaging, automotive, electrical, agriculture, etc.).

Among these three grades of PE; LDPE is the most widely used PE grade, due to its relatively good mechanical and optical properties, combined with a competitive market price [4]. LDPE is known to has good properties, such as: high flexibility, good processability, impact resistance, dimensional stability and excellent resistance to dilute acids, alcohols, bases and esters [5]. In actual fact, LDPE plays a key role in the industry of PE due to its wide range of applications. On the other hand, HDPE has a stronger intermolecular forces, higher density and greater proportion of crystalline regions than LDPE. The size and size distribution of crystalline regions are determinants of its properties. HDPE is used in the production of plastic bottles, corrosion-resistant piping, geomembranes and plastic lumber. Although all PE grades are at the top of the list of commodity polymers, they are susceptible to degradation.

Polymer degradation is a change in the properties of a polymer under the influence of one or more environmental factors such as heat, light (UV radiation), mechanical stress or chemicals. Degradation could be defined as a change in the chemical structure of a plastic involving a deleterious change in properties [6]. Degradation in general may occur during processing, manufacturing, storage and end use. PE grades can absorb solar UV radiation and undergo photolytic, photo-oxidative, and thermo-oxidative reactions that result in the degradation of these materials. This could cause catastrophic failure and shorten their useful lifetime [7]. It has been stated that the hierarchy in the oxidation susceptibility of PE is as follows: LDPE > LLDPE > HDPE [8-10]. The susceptibility of a polymer to degradation depends on its structure. Basically, the susceptibility of a polymer to degradation would depend on the chemical composition, molecular weight, crosslink density, degree of crystallinity, and environmental degrading conditions [11-12].

UV radiation and oxygen are the most important factors that initiate degradation, leading to release a free radicals. Subsequently chain scission takes place. The physical and mechanical properties of the polymer deteriorate and its average molecular weight (chain length) decreases, melt flow rate increases, degree of crystallinity increases and a powdery surface eventually forms [13-15]. Moreover, degradation products such as alkanes, alkenes, ketones, aldehydes, alcohols, carboxylic acids, keto-acids, linear esters and lactones could be formed [16]. However, studying the degradation process of polymers is crucial. The usefulness of any material, including polymer blends, depends on its degradability and durability [17]. For example, polymers which used as a building materials, especially in outdoor applications should be highly resistance to degradation. This is because it is difficult and expensive to repair or replace them. The main techniques used to evaluate the polymer degradation could be divided into surface analysis (infrared spectroscopy, optical microscope, scanning electron microscope, contact angle measurements, etc.), and techniques to study changes in bulk analysis (molecular weight, degree of crystallinity, weight loss, etc.) such as gel permeation chromatography, differential scanning calorimetry, thermogravimetric analysis [18].

Blending technology could be used to either enhance the resistance of polymer to UV degradation or to increase their susceptibility towards UV degradation. According to many studies [12, 19], for most of the polymer materials, blending is the first choice and a classic way to enhance their UV resistance. The resistance of polymers such as PE to UV degradation could be enhanced by the use of additives which absorb the UV radiation (e.g. stabilizers or blockers), or by coating or blending with another polymer which is opaque to UV radiation. Other studies [16-20] claimed that the susceptibility towards degradation could be enhanced by introducing UV absorbing groups along the PE chain or by blending PE with UV absorbing additives or blending with functional polymers and compounds. Other properties such as physical, mechanical, electrical, rheological is known to improve by blending [21-22]. Therefore, the major objective of this study is to investigate the effects of blending composition on the UV stability of LDPE/HDPE blends. On the basis of the above facts and information, it seems possible to play with the UV stability of PE and their blends by blending one grade of PE such as LDPE with other PE grade such as HDPE. In a previous study [23], mechanical properties of LDPE/HDPE blends were investigated. The results of this study demonstrated that the mechanical properties of LDPE/HDPE blends were varied according to HDPE content. Consequently, gathering the results of the current study with the previous ones would provide more understanding and insights about the properties and applications of these blends associated with their composition.

## Experimental

### Materials

The polymers used in this study are commercial grades; LDPE from Lyondel Basell (PE 1840H) and HDPE from Ras Lanuf Co. (Libya) (HDF-5116). The melt flow index (MFI) and density values of LDPE are 1.5 g/10min (ASTM D 1238, 190 °C, 2.16kg) and

0.919 g/cm$^3$ (ASTM D 1505, 23 °C), respectively. LDPE (PE 1840H) can be used for blow-fill-seal applications such as bottles, vials and healthcare applications like caps, closures, collapsible tubes, medical devices and medical films. Whereas the melt flow index (MFI) and density values of HDPE are 0.15g/10min (ASTM D1238, 190 °C, 2.16kg) and 0.951g/cm$^3$ (ASTM D 1505, 23 °C), respectively. This type of material is suitable for general purpose of film applications such as bags, liners, barrier film, agriculture mulching film and co-extrusion film.

### Blending process

LDPE/HDPE blends with varying ratio (80/20, 60/40, 40/60, and 20/80), were prepared by melt-mixing technique using mini-twin-extruder (Haake mini CTW twin screw) with average screw speed of (90 rpm.), and the barrel temperatures set on 200 °C. The specimens for accelerated weathering test were prepared using mini-injection mould (Xplore 12ml) at melting temperature 230 °C, injection pressure (1.6 MPa), and holding time (6 s).

### Accelerated weathering test

The LDPE, HDPE and their blends were conducted and exposed to UV-B light (340 nm) for 415 h (~ 17 days of 4 periods/seasons) in accelerated weathering test machines (Teemi880, china). The test consisted of four cycles that simulates day, night, rain, and condensation according to the four seasons of the year. These cycles were started with cycle of ~ 104 h of exposure to UV light at temperature of 40±2 °C, and relative humidity of 80±5% (Summer season), followed by ~ 104 h of exposure to UV light at temperature of 23±2 °C, and relative humidity of 20±5% (Spring season), followed by ~ 104 h of exposure to UV light at temperature of 18±2 °C, and relative humidity of 15±5% (Autumn season) and eventually followed by ~ 104 h of exposure to UV light at temperature of 8±2 °C, and relative humidity of 15±5% (Winter season). Samples had a length of 73 mm, thickness of 2 mm, and wide of 4 mm.

## Recognition of degradation

### Fourier transform infrared spectroscopy (FTIR)

It is well known that the most suitable and sensible analysis for assessing polymer degradation is fourier transform infrared spectroscopy (FTIR) [24]. FTIR (Bruker, Tensor II) analysis of LDPE, HDPE and their blends was conducted using ATR-FTIR mode. All spectra were recorded from 450 to 4000 cm-1 at a resolution of 4 cm$^{-1}$.

Carbonyl index (CI) was use to estimate the degree of degradation of LDPE, HDPE and their blends before and after accelerated weathering test. Carbonyl groups' formation is an effective parameter for monitoring the photo-oxidation degree of PE and could be presented as CI [25]. The content of carbonyl groups, formed by the oxidation of PE, could be quantified by a CI obtained by dividing the stretching carbonyl band area (1718 cm$^{-1}$) by the area of methylene group symmetric stretching vibration (2830 cm$^{-1}$).

The CI of LDPE, HDPE and their blends before and after weathering was calculated according to the following equation [26]:

$$\mathrm{CI} = \frac{\text{absorption at } 1775\ \mathrm{cm}^{-1}}{\text{absorption at } 2830\ \mathrm{cm}^{-1}}$$

### Contact angle

All the measurements were carried out using Contact angle ramè-hart instrument co. model 200-F4 at room temperature. 3 μl volume drops of water were deposited on the surface of the LDPE, HDPE and their blends using a syringe. Pictures of the water drops were acquired through a digital camera positioned on a static contact angle analyzer. The θ of the contact angle was measured automatically from the image setup. Each contact angel value is an average of 10 measurements and the standard deviation were calculated.

### Optical microscope

Microscopic observations of LDPE, HDPE and their blends surface changes after exposure to UV were carried out by means of an optical microscope (XP-501 transmission polarizing microscope, Turkey), equipped with a color digital camera (Moticam 2) and software Motic Images Plus 2 at different magnification.

## Results and discussion

### FTIR analysis

Fig. 1 shows the FTIR spectra of LDPE, HDPE and their blends recorded before and after exposure to UV radiation. Fig. 1a shows that the main FTIR bands characteristics of PE (both LDPE and HDPE) are the stretching vibration of carbon-hydrogen (CH) group of the main chain at 2770-3040 cm$^{-1}$ and the wagging and rocking vibration of methylene ($CH_2$) at 1430-1490 cm$^{-1}$ and 690-750 cm$^{-1}$, respectively [27-29]. This is because LDPE and HDPE share the same major structural unit, functional groups, chemical bonds [30-31] and therefore have many identical wavelength. However, the different degree of branching results in small, but important differences. For example, FTIR spectrum of HDPE in Fig. 1a shows a single band at 700-750 cm$^{-1}$ and at 1440-1490 cm$^{-1}$ because of their linear structure [32]. These bands were appeared as two double bands in the case of LDPE. Moreover, FTIR spectrum of LDPE in Fig. 1a had a unique characteristic band (quite small) at 1375 cm$^{-1}$, representing a methyl group ($CH_3$) bending deformation of the branched chain ends. This band was absent in the case of HDPE [32]. These results indicate that even similar polymers such as LDPE and HDPE can be distinguished using FTIR spectra. As can be seen in Fig. 1b, exposure to UV radiation caused significant degradation of LDPE, HDPE and their blends. The most significant changes in FTIR spectra after exposure to UV radiation were the carbonyl (1550-1740 cm$^{-1}$), amorphous (1260 cm$^{-1}$) and hydroxyl regions (3400 cm$^{-1}$). Thus, the bands at 1715 cm$^{-1}$ for the degraded LDPE, HDPE and their blends (shown in Fig. 1b) is due to the formation of a variety of carbonyl groups, while the broadband between 3100 and 3600 cm$^{-1}$ can be assigned to the formation of hydroxyl and hydroperoxide groups [26]. The intensity of

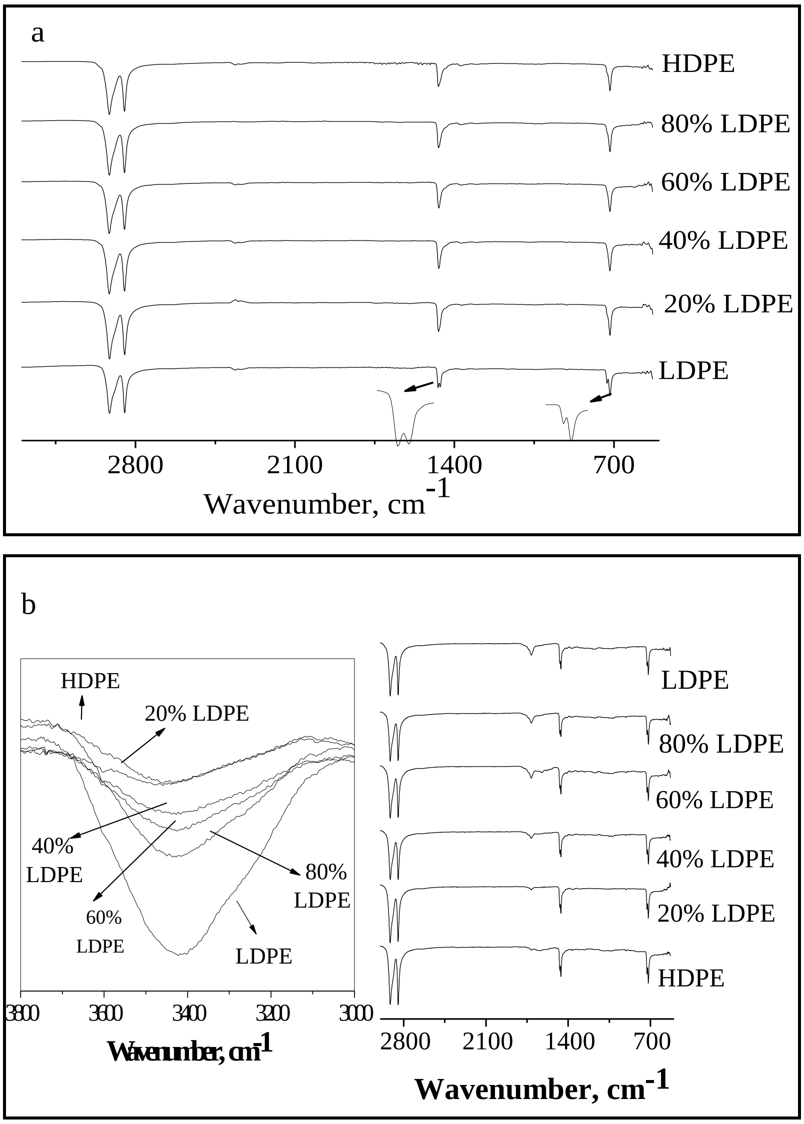

Fig. 1. FTIR spectra for a) LDPE, HDPE and their blends b) LDPE, HDPE and their blends after exposure to UV radiation.

these bands were somehow higher in the case of LDPE than in the case of HDPE, suggesting that HDPE is less susceptible to degradation than LDPE, as reported in the literature [33-35]. This is because LDPE is a branched polymer with a greater number of tertiary carbon, which makes it more susceptible to undergo photo-oxidation reactions [31]. Achilias et al. [36], cited that less crystalline or more branched polymers are less stable during the degradation process. On the other hand, the intensities of the same formed bands (carbonyl, hydroxyl and hydroperoxide groups) after exposure to UV radiation as shown in the FTIR spectra in Fig**.** 1b of LDPE/HDPE blends decreased as the amount of HDPE were increased in the blends, which indicate that the degradation was decreased with increasing the amount of HDPE in the blend composition.

The CI of LDPE, HDPE and their blends before and after weathering was calculated as shown above. According to Chiellini et. al. [33], carbonyl groups usually account for most of the products of thermo-oxidative degradation of PE, their concentration, as determined by the CI, could be used to detect the degradation. Generally, a higher value of CI indicates a higher degree of polymer degradation [25]. Before exposure to UV radiation, CI of LDPE were higher than that of HDPE as shown in Fig. 2. As mentioned above, degradation could be introduced to the polymer during, manufacturing (e.g. polymerization), processing (e.g. extrusion), storage and use [4]. Also, the results in Fig. 2 illustrates that the CI of LDPE, HDPE and their blends before exposure to UV radiation were decreased as the HDPE content increased in the blend. This properly indicate that the resistance of LDPE/HDPE blends to degradation was increased as the HDPE content increased in the blend. Furthermore, it can be seen that, CI was increased after weathering test (after exposure to UV radiation) for LDPE, HDPE and their blends. It can be also observed that, even after weathering the resistance to degradation was increased as the HDPE content increased in the blend.

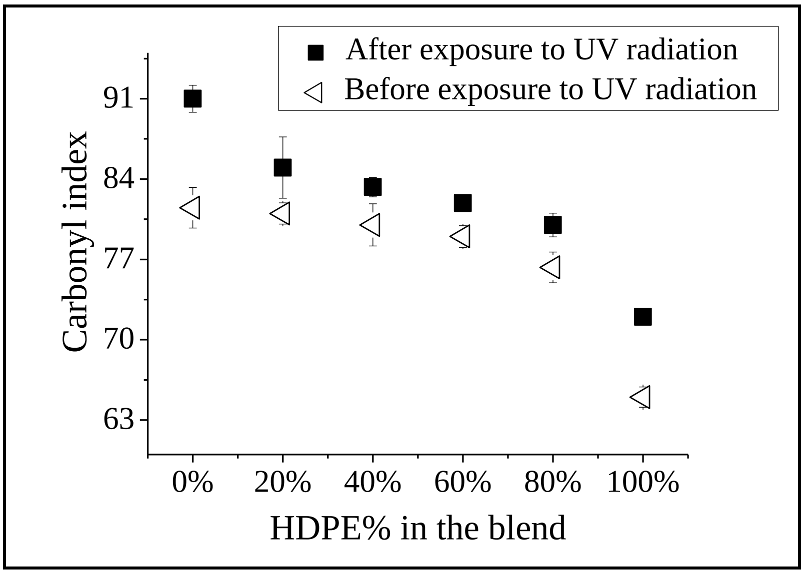

Fig. 2. Carbonyl index of LDPE, HDPE and their blends before and after exposure to UV radiation.

## Contact angle

It should be pointed out that obtained results characterize degradation mostly on the surface of the LDPE, HDPE and their blends, while the degradation is supposed to be significantly less present in polymer bulk [25]. As a matter of fact, degradation is associated with chemical and physical changes taking place on the surface of polymer [37]. For this reason, contact angle measurement (CAM) was performed in this study to follow the degradation and detect the structural changes on LDPE, HDPE and their blends surfaces. Ordinarily, the value of the contact angle can vary from 0-180$^o$, 0$^o$ representing fully wetted surfaces and 180$^o$ representing totally non-wettable surfaces [38]. Additionally, hydrophobic surfaces are characterized by a contact angle of 90$^o$ or more and hydrophilic surfaces are characterized by a contact angle of less than 90$^o$. In practical, wettability and hydrophilicity are closely related phenomena. More wettability means more hydrophilicity. However, CAM depends on several factors, such as surface energy, wettability of the surfaces, viscosity of the liquid, roughness, the manner of surface preparation, and surface cleanliness [39-40].

The values of the contact angle for LDPE, HDPE and their blends before and after exposure to UV radiation are shown in Table 1. Standard deviations are given in parentheses. The values of the contact angle for LDPE, HDPE and their blends before exposure to UV radiation were varied from 73.2$^o$ (for LDPE) to 91.3$^o$ (for HDPE). Normally, LDPE and HDPE are inherently hydrophobic polymers due to their non polar surfaces. The hydrophilicity of LDPE could be due to the degradation, which may occur during processing, manufacturing, and storage. It is important here to emphasis that the resistance of LDPE to UV degradation improves by blending with HDPE and vice versa.

The values of the contact angle for LDPE, HDPE and their blends after exposure to UV radiation were varied from 70.5$^o$ to 83.3$^o$, as shown in Table 1. Exposure to UV radiation caused decrease in the contact angle for LDPE, HDPE and their blends, as shown in Table. This is obvious because the radiation under UV light increases the hydrophilicity of PE surface by introducing of certain polar groups, as stated above. This means that the surface of HDPE and their blends became hydrophilic.

Table 1. Contact angle and standard deviation of LDPE, HDPE and thier blends before and after exposure to UV radiation.

| Sample | Contact angle, ɵ | |
|---|---|---|
| | Before exposure to UV radiation | After exposure to UV radiation |
| LDPE | 73.2$^o$ (1.1) | 70.5$^o$ (3.6) |
| 80%LDPE | 85.4$^o$ (3.6) | 79.1$^o$ (3.2) |
| 60%LDPE | 85.6$^o$ (3.6) | 79.8$^o$ (3.1) |
| 40%LDPE | 85.8$^o$ (3.4) | 80.2$^o$ (0.9) |
| 20%LDPE | 86.0$^o$ (1.3) | 83.3$^o$ (3.1) |
| HDPE | 91.3$^o$ (2.8) | 79.9$^o$ (0.7) |

## Optical microscope

Surface degradation could be also investigated by optical microscope [41]. Optical micrographs for LDPE, HDPE and their blends after exposure to UV radiation are shown in Fig. 3. These micrographs support the FTIR and CAM results. More cracks were present in LDPE than in HDPE. These cracks were decreased with increasing the amount of HDPE in the blend composition. This once again indicates that degradation was decreased with increasing the amount of HDPE in the blend composition.

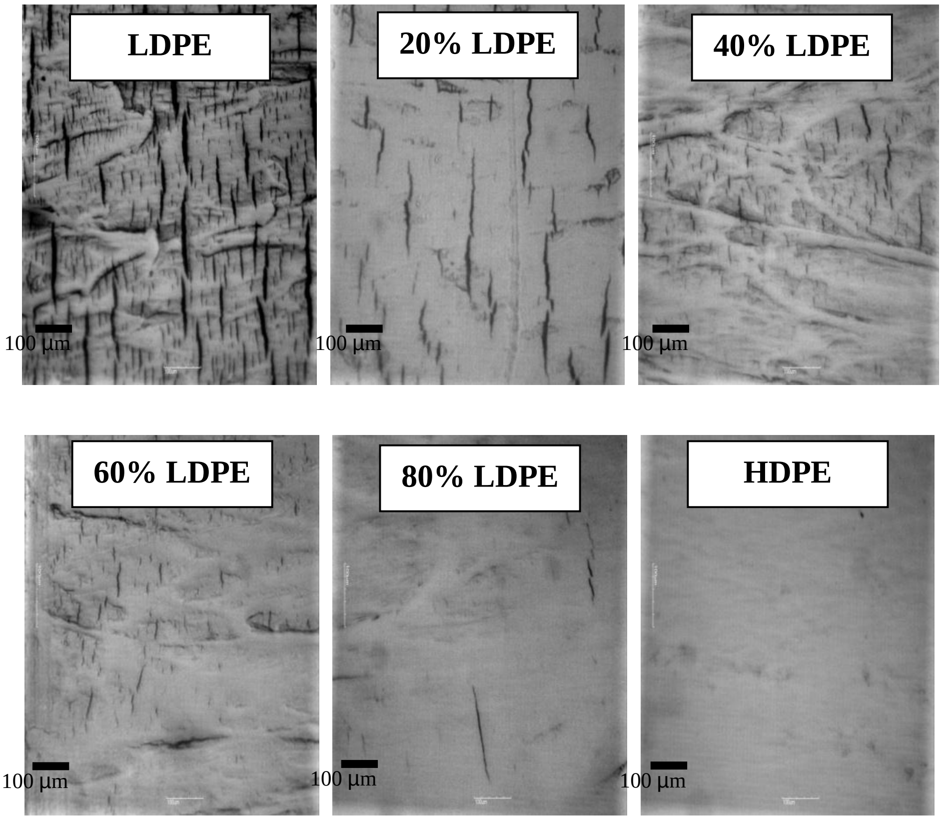

Fig. 3. Optical micrographs of LDPE, HDPE and their blends after exposure to UV radiation.

Gathering the findings of this study with previous ones (reference 23) could be useful in many ways:

- Blending composition could be used as indicator to predicate the final properties and suitable application of the final LDPE/HDPE blend.
- UV stability of the LDPE/HDPE blends depends on its chemical composition. For example, blends-rich with HDPE provide better resistance to UV radiation, while blends-rich with LDPE can be more eco-friendly and posses more degradability. This would enable industrial and users to identify and select appropriate blends for specific environments, particularly where durability issues are of significance.
- Resistance of LDPE to UV degradation could be improved by blending with HDPE. This would enable industrial and companies to identify and select appropriate LDPE/HDPE blends to replace LDPE for certain outdoor applications, particularly where the exposure to UV radiation issue are of significant.
- The desired properties for the final LDPE/HDPE blend to be used in suitable application could be predicated based on the composition. For example, the resistance to UV degradation and overall mechanical properties gives the blend with the composition of LDPE (60) / HDPE (40) considerable attention to be used in outdoor

pplications. Blend (e.g. LDPE (80) / HDPE (20)) possess a light weight and good toughness which may be used for creating a wide range of goods that include household plastic products, automotive interior and exterior components and a number of biomedical devices. Blends-rich with LDPE can replace LDPE to make films which suitable for a host of applications, such as mulching, packaging of consumer goods, and industrial products such as fertilizers. On the other hand, blends-rich with HDPE can replace HDPE in certain applications such as manufacture of heavy duty films for fertilizers, planting and merchandise bags and containers.

## Conclusion

The effects of blending composition on the UV stability of polymer blend comprising different grades of PEs were investigated. This investigation was accomplished by studying the susceptibility of LDPE/HDPE blends with varying ratio (80/20, 60/40, 40/60, and 20/80) to UV degradation using accelerated weathering test. After weathering test, FTIR results indicated the formation of hydrophilic groups such as carbonyl, vinyl, and hydroxyl in the LDPE, HDPE and their blends. CI was increased after weathering test for LDPE, HDPE and their blends, confirming the formation of the hydrophilic groups. The formation of the hydrophilic groups caused reduction in the contact angle values and wettability changes for LDPE, HDPE and their blends. The optical microscopic observation results showed visual evidence of surface and morphological changes after weathering test for all the polymers and blends. It is very important to conclude here that blending composition plays substantial role in defining the degradability of the blend. The susceptibility of LDPE/HDPE blends to UV degradation appeared to decrease with increasing the amount of HDPE in the blend composition.

In our previous work, it was reported that the mechanical properties such as tensile strength and hardness showed to increase with increasing the amount of HDPE in the blend composition. In line with previous study, and based on the results obtained in this work, it is important to conclude here that the resistance of LDPE to UV degradation could be improved by blending with HDPE without substantial loss in its properties. Moreover, blend composition can be optimized to meet specific end use property requirements. In other words, blending composition can be used as indicator to predicate the final properties and suitable application of the final LDPE/HDPE blend.

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