

Photooxidative behaviour of polyethylene/polyamide-6 blends

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ARTICLE INFO

Article history:

Received 8 December 2009

Accepted 24 December 2009

Available online 11 January 2010

Keywords:

Photooxidation behaviour

Polymer blends

Mechanical properties

Infrared analysis

ABSTRACT

The photochemical behaviour of several polyethylene/polyamide-6 blends was studied under conditions of artificial accelerated weathering. Particular attention was paid to five different compositions ranging from pure polyethylene to pure polyamide with blends of PE/PA-6 of various compositions: 75/25, 50/50 and 25/75 wt/wt%. Analysis by infrared spectroscopy of the chemical modifications caused by photooxidation showed that exposing the polyethylene/polyamide-6 blends to UV-light irradiation led to the formation of oxidation photoproducts in both polymer phases. In agreement with both the mechanical and spectroscopic analyses, the photooxidation rate of the blends was observed to be much higher than that of the homopolymers. The results given in this paper suggest that photooxidation of the PE/PA blends starts in the polyamide phase and that the subsequent photooxidation of the polyethylene phase may be initiated by the radicals coming from the oxidation of PA.

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1. Introduction

In recent years, polymer blending has been developed as a simple method for tailoring the properties of polymeric materials without investing in new chemistry. Among the many immiscible polymer blends of technological interest, we have focused our attention on blends of low density polyethylene and polyamide-6, which are widely used in a variety of packaging and automotive products due to their good processability, high chemical barrier properties and mechanical properties. Polyethylenes offer high impact strength resistance and they serve as good barriers to moisture, while polyamides have good mechanical properties and have very low oxygen permeability. Unfortunately, low density polyethylene and polyamide-6 are immiscible and form phase separated blends, with the properties of these blends not as good as predicted using a continuum model. In order to improve the macroscopic properties of these blends, it is necessary to use a precursor as a compatibiliser, such as maleic anhydride grafted polymers or ethylene/acrylic acid copolymers [1–3].

Whatever the application, the durability of polymeric materials is always important, in order to have adequate lifetimes, maintenance and replacement. Material degradation is the deterioration in

the properties of the polymer that takes place due to different factors of the environment. As a consequence of degradation, the life of the product diminishes. Thus, any polymeric material that will be used in outdoor applications where durability is a requirement should be highly resistant to the environmental conditions. For outdoor applications of polymers such as packaging films and automotive products, it is peculiarly important to insure adequate resistance to photooxidation. The exposure of products to UV light provokes photooxidation, which manifests as modifications in the chemical structure of the macromolecular chains, the variation of molecular weight, and changes in a material's morphology, all of which lead to a decrease in the macroscopic properties [4–7]. There are only limited papers in the literature that deal with the photooxidation of polymer blends. These papers are mainly concerned with photodegradation of blends from same polymer, in virgin and recycled forms (monopolymer blends [6]), incompatible blends [8] or the blends from compatible or semi-compatible polymers [9–12].

The photooxidation of polymer blends is a complex phenomenon, since both components are susceptible to oxidation. Radical species can be formed in the two homopolymers, and these species can interact. Therefore, the complete oxidation mechanism may be a combination of the oxidations of the two components. Moreover, the diffusion of oxygen should be taken into account in the characterisation of the mechanism, since it can largely differ from one of the component to the other.

This work reports a study of the photooxidation of several PE/PA-6 blends with different PE/PA-6 ratios. The samples were exposed to conditions of artificial accelerated weathering. The

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photooxidation behaviour was analysed by infrared spectroscopy and the mechanical properties were characterized. This study aims to provide a better understanding of the UV-light oxidation of polymer blends.

2. Experimental

2.1. Materials

The materials used in this work were a low density polyethylene (PE) (Riblene FC30, from Polimeri Europa, Italy), $M_w = 175,000 \text{ g mol}^{-1}$, $M_w/M_n = 5.76$, $MFI_{190^\circ\text{C}/2.16\text{kg}} = 0.28 \text{ g } 10 \text{ min}^{-1}$, $\rho_{23^\circ\text{C}} = 0.922 \text{ g cm}^{-3}$ and a polyamide (PA-6) (Radilon® S, from RadiciGroup, Italy), with an intrinsic viscosity of 2.05 dL g^{-1} (measured in formic acid 80% vol/vol at 30°C), and a density $\rho_{23^\circ\text{C}} = 1.14 \text{ g cm}^{-3}$.

2.2. Processing and characterisations

Three different blends with compositions 75/25, 50/50 and 25/75 PE/PA wt/wt% were made. The samples (including the two homopolymers and the three blends) were made in the form of thin films. The blends were prepared by melt mixing the homopolymers in a Brabender Plasticorder mixer model PLE 330, at 240°C , with a stirring speed of 60 rpm. The samples were mixed for approximately 10 min, which was long enough to obtain a constant torque value. For consistency, the two homopolymers were subjected to the same processing conditions. Films were obtained by compression moulding of the materials after the mixing, with a final film thickness of $50 \mu\text{m}$.

The polymer films used for the characterisation of the mechanical properties were irradiated in a Q-UV chamber ($\lambda > 300 \text{ nm}$, 8 h of light at $T = 60^\circ\text{C}$ and 4 h of condensation at $T = 40^\circ\text{C}$). The samples used for the characterisation by infrared spectroscopy were exposed in a SEPAP 12.24 unit [13] ($\lambda > 300 \text{ nm}$ in the presence of air, R.H. = 2%, temperature = 60°C).

Infrared spectra were recorded with a Nicolet 5SX-FTIR spectrometer using OMNIC software. Final spectra were obtained from the summation of 32 scans with a resolution of 4 cm^{-1} . To compare the photooxidation rates of the different formulations, it was necessary to normalize the spectra. A correction to the absorbance measured by infrared spectrometry was applied to the raw data. When one considered the photoproduct attributed to PE, the absorbance was corrected according to the following equation: $A_{\text{corrected}} = A \times 100/(\%PE)$. The same correction was used for a photoproduct attributed to PA-6: $A_{\text{corrected}} = A \times 100/(\%PA-6)$.

The mechanical tests were performed using an Instron machine model 4443 on samples cut from the compression-moulded films.

3. Results and discussion

3.1. Mechanical properties

First, the mechanical behaviour, in particular the tensile strength and the elongation at break, of all the PE and PA-6 blends was monitored as a function of their composition. Fig. 1 shows the tensile strength and the elongation at break values for the non-irradiated samples as a function of polyamide content.

The results indicate that the tensile strength values of the blends are between those of the two homopolymers, but their values are lower than those expected on the basis of a simple additive rule. The elongation at break as a function of the polyamide content for the non-irradiated samples shows a more complex behaviour. In particular, all the values are not only less than predicted from an additive rule, but there is a deep minimum observed between

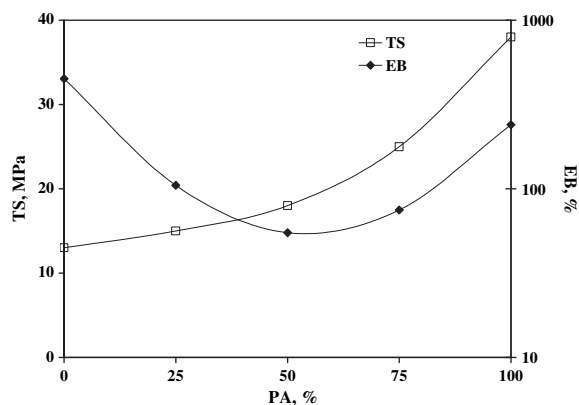


Fig. 1. Tensile strength and elongation at break of all non-irradiated blends as a function of their composition.

compositions PE/PA 50/50 and 25/75 wt/wt%. This indicates an antagonistic effect due to the lack of interactions between the two polymeric phases. In the absence of the compatibiliser precursor, the antagonism between the two phases is a determining factor for the morphology and therefore the properties as well [2,3].

Fig. 2 shows the dimensionless tensile strength of all the investigated samples, reported as a function of the irradiation time in the Q-UV unit. The dimensionless values were calculated as the values of tensile strength at a given irradiation time, divided by the corresponding values for the non-irradiated sample.

It can be observed that the tensile strength decreases slowly with the exposure time for the pure polyethylene and much more rapidly in the case of the pure polyamide. Increasing the amount of PA in the blend results in a decrease of the tensile strength that is near linear for all the irradiation times. However, one can notice an antagonistic effect in the case of the 25/75% blend, which shows a larger decrease of the relative TS value than the pure PA sample.

Much more impressive is the effect of photooxidation on the changes of the dimensionless elongation at break (EB), as shown in Fig. 3. Indeed, the elongation at break is more sensitive to the molecular and structural variations and to the morphology of the material. Here again, the values given in the figure are normalized to the initial value before irradiation.

The two homopolymers, especially the pure polyethylene, show only a slight decay of the elongation at break after short exposure times. For longer times of exposure, one can observe a dramatic decrease of the EB values for both samples. If one compares these

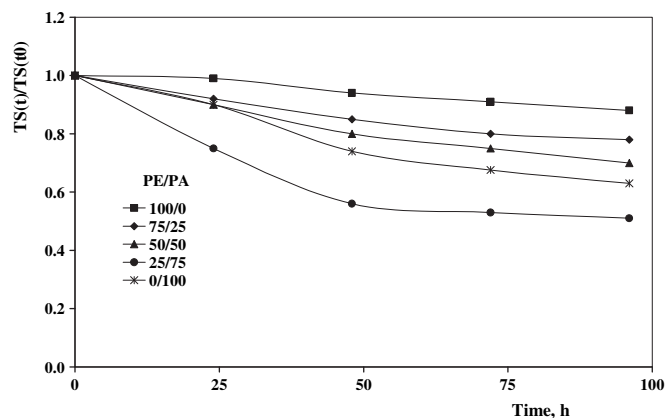


Fig. 2. Dimensionless tensile strength as a function of the UV-B irradiation time of the pure polyethylene and polyamide and all the blends.

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