

# Artificial and Natural Weathering of ABS

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**ABSTRACT:** Acrylonitrile-butadiene-styrene (ABS) is a complex copolymer with exceptional importance in technological applications. However, ABS is known to present a high susceptibility to weathering when exposed to UV radiation in the presence of oxygen. In this work, photodegradation of ABS was studied under natural and accelerated conditions. Exposed samples were removed periodically and characterized by several analytical techniques. During both types of weathering chemical modifications occurs mainly at the surface, which affects

considerable the optical, mechanical, and rheological properties. The results obtained evidence that during accelerated weathering the formation rate of photodegradation products is higher when compared to natural weathering results. However, chain scission occurs mainly along natural conditions. © 2010 Wiley Periodicals, Inc. *J Appl Polym Sci* 116: 2005–2014, 2010

**Key words:** ABS; photodegradation; yellowness; mechanical properties

## INTRODUCTION

Acrylonitrile-butadiene-styrene (ABS) is a styrenic copolymer with a wide range of applications in plastic industry due to their excellent combination between cost, convenient manufacturing conditions, and mechanical, thermal, chemical, and electrical properties.<sup>1</sup> This material exhibits an important particularity because its properties can be modified by changing grafting conditions and monomer amount for specific applications.<sup>2</sup> ABS is used in outdoor applications, where good resistance to UV radiation is required. However, this material is known to present weathering instability, resulting in properties loss. These changes can be followed by chemical modifications, such as crosslinking and chain scission, color changes, and mechanical properties.<sup>3–6</sup>

There are several factors that can lead to degradation, namely external causes related with environmental conditions (UV radiation, oxygen, moisture, and temperature) and inherent polymer features (free volume, glass transition temperature, morphology, and impurities).<sup>6</sup>

The work that have been carried out on ABS degradation showed that ABS copolymer is very suscep-

tible to photo-oxidative degradation due to the presence of butadiene.<sup>7–16</sup> This instability is related to the labile position of the hydrogen on the carbon atom in  $\alpha$ -position of the carbon-carbon double bonds that can be easily abstracted.<sup>17</sup> Piton et al.<sup>18</sup> studied the photo-oxidation of ABS thin films (100  $\mu\text{m}$ ) at long wavelengths ( $\lambda > 300 \text{ nm}$ ) in the presence of oxygen and the results obtained were compared with polybutadiene and polystyrene homopolymers. The results showed that the butadienic component acts as a photoactivator, inducing oxidation of the polystyrene component in the styrene-acrylonitrile (SAN) macrophase. Gardette and co-workers<sup>19,20</sup> showed that degradation of ABS films, irradiated at  $\lambda > 300 \text{ nm}$ , occurs at the surface and its extension depends on the sample thickness. Recently, Schlick et al.<sup>21</sup> studied ABS films (4 mm) containing a hindered amine stabilizer exposed to radiation in the range of 290–330 nm using ATR-FTIR. The results demonstrated that degradation was a heterogeneous phenomenon and the surface exposed directly to radiation undergoes larger damages when compared to inner layers, in agreement with observations made by other authors.<sup>19,20</sup>

Most of the scientific research until now, on photochemical processes occurring during ABS degradation, was performed under accelerated conditions. The development of methods allowing acceleration of the degradation processes during films lifetime was considered very important. Several equipments have been used in accelerated aging tests (Atlas Weatherometer Ci 3000, Atlas Uvcon, Atlas XR 260 weatherometer, Xenotest Type 450, Suntester, QUV

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and Sepap) to predict the polymer lifetime under service conditions.<sup>6</sup> The correlation between accelerated and natural weathering is not trivial and depends of many agents: accelerated weathering devices, geographical localization in natural experiments, temperature, amount of sunshine hours, mechanical stresses, biological attack, and environmental contaminants.<sup>20</sup>

It is known that a good correlation between different weathering devices is found when the equipment operates with the same type of radiation source and under similar conditions, being the temperature an important parameter.<sup>22-27</sup>

Recently, Pickett et al.<sup>26</sup> studied the effect of the irradiation conditions on the weathering of engineering thermoplastics, such as polycarbonate (PC) and blends of PC with other polymers, such as ABS and SAN. Aiming to establish the authenticity of the accelerated tests to reproduce the effects of prolonged exposure to natural weathering, the influence of several parameters, namely light intensity, spectral distribution, light/dark cycle, and different filter conditions (boro/boro, CIRA/soda, and quartz/borosilicate) were investigated. The results demonstrated that the samples submitted to shorter wavelength suffer more damages. Moreover, for ABS a nonlinear correlation between irradiance and degradation was obtained.

Even though, lot of studies on polymer degradation have been performed under accelerated conditions, it is essential to reproduce the natural weathering phenomena to evaluate the real durability of these materials.<sup>16,17,28,29</sup> Thus, this work aims to obtain and compare the degradation process occurring during artificial and natural weathering of ABS. Experiments were carried out to obtain details on the influence of ultraviolet radiation on chemical structure and physical properties. Nonstabilized ABS samples were submitted to natural and accelerated weathering. As filtered Xenon lamp present an UV spectrum comparable to UV spectrum of the sun and this pattern of light sources, at low temperature, is expected to be representative of outdoor aging,<sup>30,31</sup> hence it was selected.

## EXPERIMENTAL

### Material

Experiments were performed on unstabilized ABS (Terluran HI-10 from BASF) kindly supplied by Colorgal-Cores de Portugal, Lda. (Lisbon, Portugal). ABS Terluran is a high impact grade with medium flow rate suitable for injection molding and extrusion.

### Sample preparation

Polymer granules, previously dried in an oven at 80°C during 1 h, were extruded in a "Luigi Ban-

**TABLE I**  
**Tape Extrusion Conditions**

Settings	Temperature (°C)
Barrel zone #1	200
Barrel zone #2	200
Barrel zone #3	200
Barrel zone #4	190
Extrusion die	175
Melt	185

dera" single-screw extruder (L/D 30) under specific processing conditions (Table I) to produce tape (0.5 mm thickness). The screw speed used was 75 rpm.

The extruded tape was cooled in water at a constant temperature, of approximately 40°C and dried with static air. Later, the tapes were cut according to required dimensions and submitted to natural and accelerated weathering, in agreement to standard methods.

### Photodegradation experiments

#### Accelerated weathering

The accelerated weathering was carried out in a Xenotest 150S chamber from Heraeus (Original Hanau) equipped with a filtered Xenon lamp with an intensity of 60 Wm<sup>-2</sup> according to standard procedures.<sup>32</sup> The light of the xenon lamp was filtered under 300 nm with an UV window combined with six IR filter glasses (according to Fig. 1).

The samples (100 mm × 2.5 mm × 0.5 mm), cuted from the extruded tape, were exposed to different degradation time period. During 2 h of aging cycle, the specimens were exposed to 18.0 ± 0.5 min of rain period and the time remaining, 102 ± 0.5 min, was considered dry period. Table II shows the conditions used in the accelerated weathering device.

#### Natural weathering

The natural weathering was performed during 4 months in Lisbon (hot, Mediterranean climate). Samples obtained under the same processing conditions as stated before, were placed in a wood support, at an angle of 45°, facing south.<sup>33</sup> Total exposure period was 4 months starting from 9<sup>th</sup> November 2007. The surface temperature was always measured using a thermocouple, being 23, 17.5, 20, and 14°C in November, December, January, and February, respectively. The average relative humidity during outdoor experiments was 73%. Table III shows more details related to monthly average values of maximum and minimum ambient temperature, relative humidity, rain, and UV exposure.

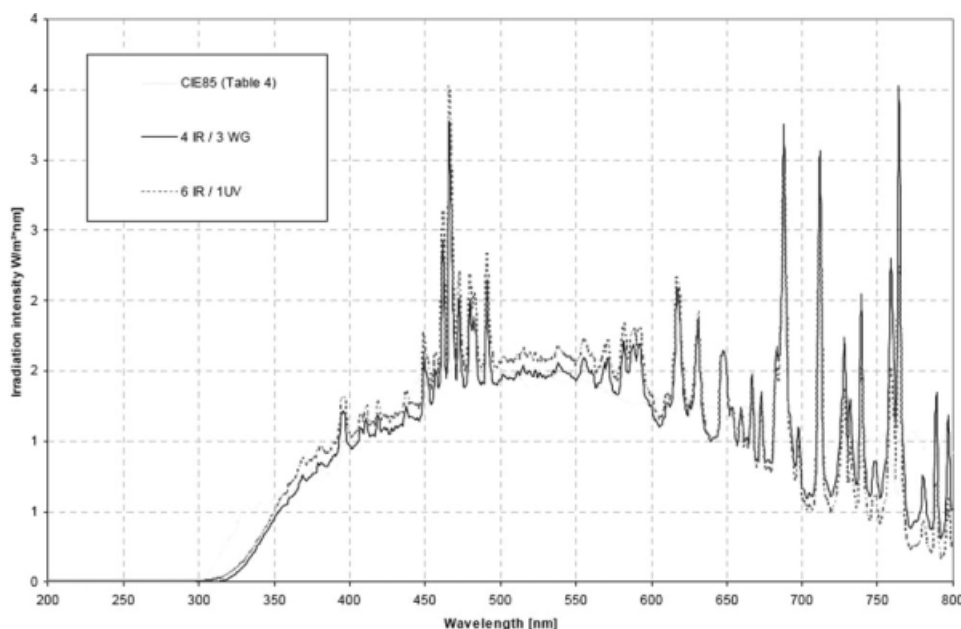


Figure 1 Spectral energy distribution in wavelength range of 200–800 nm.<sup>32</sup>

Samples were removed periodically and characterized by several analytical techniques.

### Material characterization

#### ATR-FTIR spectroscopy

Chemical modifications that occur on material surface were followed by infrared spectroscopy. All spectra (64 scans, nominal resolution of  $4\text{ cm}^{-1}$ ) between  $4000$  and  $650\text{ cm}^{-1}$  were acquired with an attenuated total reflectance accessory (ATR), using as reflecting element a ZnSe crystal using a Perkin Elmer Spectrum Spotlight 300. All spectra were aligned at  $2000\text{ cm}^{-1}$ , which corresponds to a non absorbance wavelength.

#### Color measurements

Color measurements were performed using a Data-color 650 TM spectrophotometer, light source used was D65 and the viewing angle was  $10^\circ$ .<sup>34</sup>

Color changes in the samples exposed to natural and accelerated weathering were monitored by CIE tristimulus values ( $L^*$ ,  $a^*$ , and  $b^*$ ), according to the CIELAB color system.

CIELAB is a scale that can be used to specify color changes and/or tolerances for the material through the eq. (1)

$$\Delta E = (\Delta L^{*2} + \Delta a^{*2} + \Delta b^{*2})^{1/2} \quad (1)$$

Where the coordinates represent:

$\Delta E$  – color difference

$L^*$  – luminosity in a scale from 0 to black and 100 for white.

$a^*$  – ratio between red and green, assuming positive values for the red color and negative for the green.

$b^*$  – ratio between yellow and blue, assuming positive values for the yellow and negative for the blue.

#### Mechanical testing

The mechanical properties of the ABS films nonexposed and submitted to accelerated and natural weathering were characterized using stress–strain experiments in tensile mode in a Miniature Materials Tester (MINIMAT) from Polymer Laboratories.

The tensile experiments were carried out with a deformation rate of  $0.2\text{ mm/min}$  in the direction perpendicular to the machine orientation, at room temperature. Six specimens of each same sample were tested. The thickness of every specimen was measured with a pachymeter Mitutoyo with an accuracy of  $0.025\text{ mm}$ .

#### Rheological measurements

The rheological behavior of the samples was characterized using an oscillatory rheometer TA

TABLE II  
Conditions in Accelerated Weathering Devices

Cycle period	Temperature ( $^\circ\text{C}$ )	Relative humidity (%)
Rain	23.0	85.0
Dry	30.0	58.0

**TABLE III**  
**Environmental Conditions in Natural Weathering Experiments (Average Values)**

Year/month	Maximum ambient temperature (°C)	Minimum ambient temperature (°C)	Relative humidity (%)	Rain (mm)	UV exposure (h)
2007					
November	19.1	10.3	59.6	10.8	220
December	14.4	7.8	74.5	6.0	180
2008					
January	16.4	9.7	80.6	7.0	140
February	17.2	10.2	77.8	15.8	150

instruments AR-G2 with stress-controlled fitted with plate–plate geometry with 25 mm diameter and 0.5 mm thickness. Frequency sweep was carried out to determine the complex viscosity ( $\eta^*$ ) and elastic ( $G'$ ) modulus. The experiments were performed at approximate extrusion temperature, 200°C, in a frequency range from 0.1 to 100 Hz.

#### Thermal measurements

The thermal behavior of the ABS films was evaluated by Differential Scanning Calorimetry using a Mettler DSC 821, under a constant argon flow. The samples were heated from  $-60$  to  $150^\circ\text{C}$  using a heating rate of  $20^\circ\text{C}/\text{min}$ . Three specimens of each sample were tested.

## RESULTS AND DISCUSSION

### Photodegradation products analysis

ATR-FTIR is an important tool usually used to follow the photo-oxidative degradation of polymers. This characterization technique allows to identify the structural changes occurring throughout degradation and to study the kinetic process involved in natural and accelerated weathering.<sup>35</sup>

Figures 2 and 3 present the ATR-FTIR spectra obtained for ABS samples submitted to accelerated and natural weathering, respectively. The spectra of nondegraded ABS shows C–H stretch for hydrogen atoms linked to carbons of the 1, 2 butadiene at  $911\text{ cm}^{-1}$  and of 1,4 butadiene at  $966\text{ cm}^{-1}$ . The relative absorption bands of the styrene component can be observed at  $1494$  and  $1600\text{ cm}^{-1}$ , attributed to carbon double bonds from the aromatic group. All degraded samples show a growth of the absorption bands at  $1600$ – $1800\text{ cm}^{-1}$  and at  $3000$ – $3400\text{ cm}^{-1}$ , corresponding to the carbonyl and hydroxyl groups, respectively. A decrease of the bands related to butadiene component can also be noticed.

Figure 2(a) (hydroxyl region) shows two absorption bands with maxima at  $3397$  and  $3197\text{ cm}^{-1}$ ,

which increase along degradation time. Piton et al.<sup>18</sup> identified the same photoproducts formed in ABS samples submitted to radiation at long wavelength by infrared spectroscopy with chemical treatments. The increase of absorption bands related to hydroxyl groups was attributed to alcohol and acid groups formation.

Figure 2(b) (carbonyl region) depicts a broad absorption band with a maximum at  $1646\text{ cm}^{-1}$  and shoulders at  $1600$  and  $1632\text{ cm}^{-1}$ . The shoulders of all spectra present a similar evolution that becomes considerable at 22 h. These groups result from  $\alpha,\beta$ -unsaturated hydroperoxides that can originate (by homolysis) different species, namely oxygenated compounds, such as, saturated carboxylic acids, aliphatic esters,  $\alpha,\beta$ -unsaturated aldehydes, saturated ketones, and  $\gamma$ -lactones.<sup>18</sup>

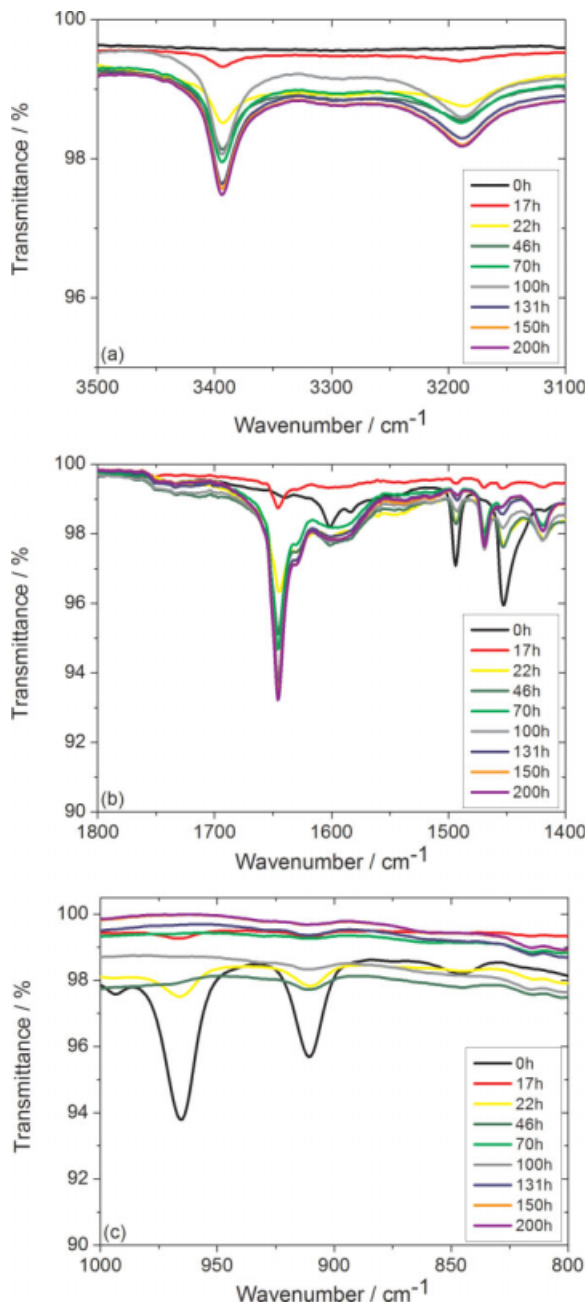
Figure 2(c) shows the butadiene region, where it is possible to observe the disappearance of the two bands at  $911$  (1, 2 PB) and  $966\text{ cm}^{-1}$  (1, 4 PB) after 70 and 46 h of exposure, respectively. This can be associated to the high reactivity of carbon double bonds of the butadiene component.

Figure 3 depicts the spectra of the samples exposed to natural weathering. The comparison of the products formed during natural and artificial weathering evidence that a similar behavior of chemical changes occurred at the same wavelengths. This suggests that the products formed during natural and artificial weathering are the same.

Using the data of the spectra of Figures 2 and 3, plots of Figures 4–6 were made. Figure 4, relative to hydroxyl region, shows two stages: the first is characterized by a fast formation of hydroxyl groups and the second characterized by the slow formation of the same groups. As seen before, this behavior can be explained by the occurrence of hydroxyl consumption in the second stage,<sup>36</sup> leading to oxide radicals and then to carbonyl groups.

A similar behavior, two stages, can be observed in the carbonyl region (Fig. 5). Under accelerated and natural aging, the formation rate of photoproducts is comparable, as it was observed by other authors for

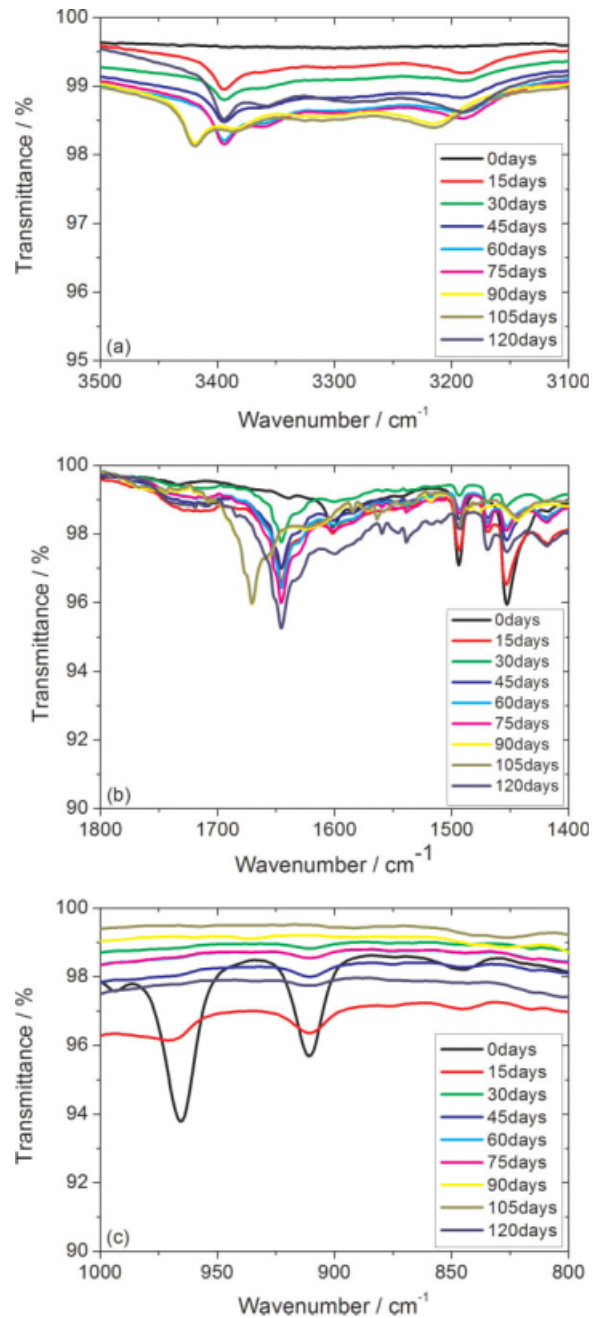




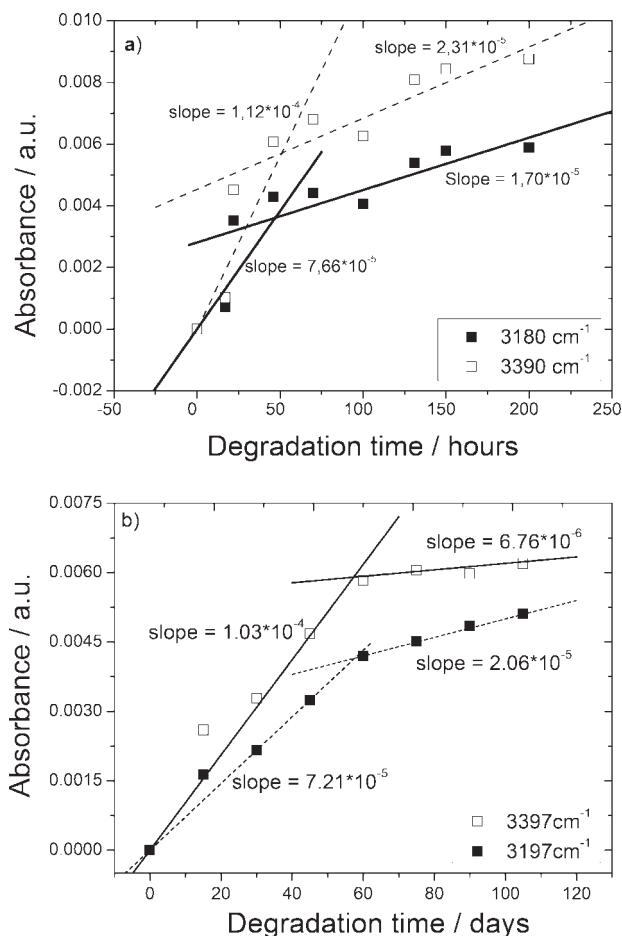
**Figure 2** ATR-FTIR of ABS films submitted to accelerated weathering: (a) hydroxyl region, (b) carbonyl region, and (c) butadiene region. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]

other polymers.<sup>37</sup> However, the results show that the chemical modifications are more pronounced in samples submitted to accelerated weathering conditions. This difference can be attributed to higher radiation intensity and consequently, higher temperature present in accelerated device, which might be associated with an increase of formation rate of degradation products.

Figure 6(a,b) show the evolution of butadiene content along the time for samples subject to accelerated and natural weathering, respectively. In both cases, a drastic decrease of butadiene component occurs in the first h/days of exposure. However, the decrease of 1, 4 PB ( $966\text{ cm}^{-1}$ ) amount is more pronounced than 1, 2 PB ( $911\text{ cm}^{-1}$ ) amount, which is in agreement with results obtained by other authors.<sup>21</sup> While



**Figure 3** ATR-FTIR of ABS films submitted to natural weathering: (a) hydroxyl region, (b) carbonyl region, and (c) butadiene region. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]



**Figure 4** Degradation rate for hydroxyl region along degradation process for ABS samples submitted to a) accelerated and b) natural weathering.

the peak at  $911\text{ cm}^{-1}$  can not be detected after 70 h for accelerated, for natural weathering it stills present before 30 days. The peak at  $966\text{ cm}^{-1}$  can not be detected after 46 h and 15 days of exposure, respectively.

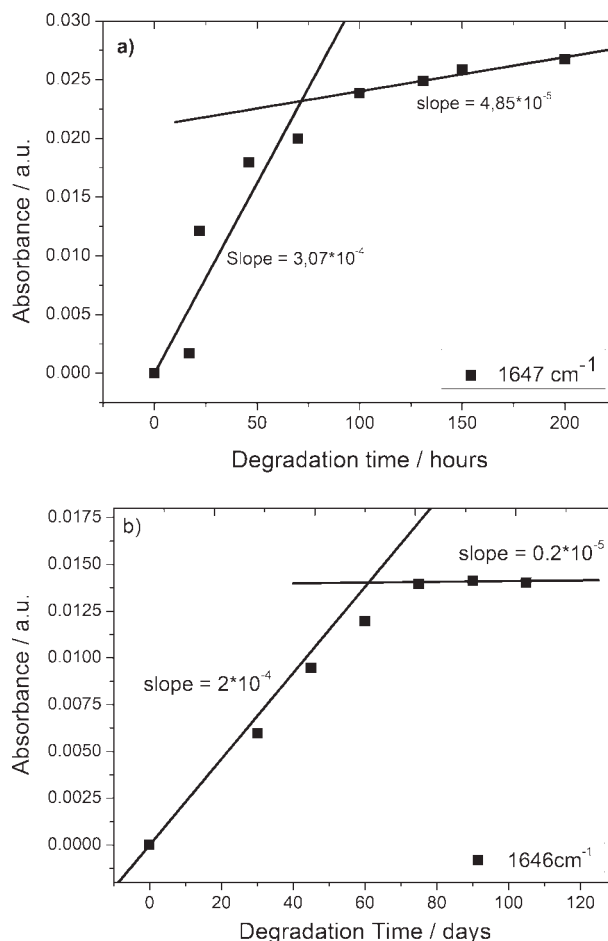
Figure 7(a,b) depict the results of the yellowness index obtained for samples submitted to accelerated and natural weathering, respectively. Figure 7(a) shows that yellowness index of the samples exposed to accelerated aging increases until 131 h. After that period of time, a constant value was reached being stable till the end of the exposure time. Similar evolution was observed for ABS films under natural aging [Fig. 7(b)], yellowness index increases during the first 60 days and tends to become stable after that period. The yellowing of the samples submitted to both types of weathering is similar. After 150 h of accelerated weathering and 105 days of natural aging, the same value is obtained, reaching approximately 28 of yellowness.

Gardette et al.<sup>16</sup> suggested that the yellowness of ABS submitted to irradiation of long wavelength in

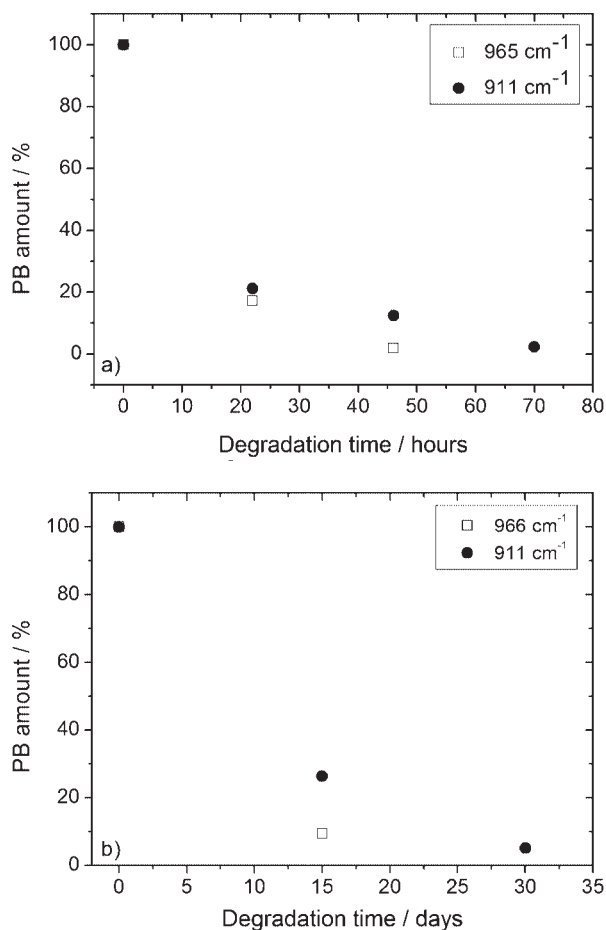
the presence of oxygen, is due to the photo-oxidation reactions of PS in the SAN macrophase, which is induced by the radicals species formed during oxidation of PB component. Due to this process, formation of many highly absorbing products (such as  $\alpha$ ,  $\beta$  – unsaturated ketones) occurs and are responsible for ABS discoloration.

### Mechanical and rheological behavior

It is known that changes in chemical and morphological polymer structure are responsible for the deterioration of mechanical properties (mainly elongation at break), cracking and disintegration of the material. To follow the mechanical behavior, stress-strain curves were obtained using ABS films with different exposure time under standard conditions. ABS is a polymeric material that exhibits a ductile behavior before exposure and as a consequence of degradation process the material becomes brittle and the plastic plateau is drastically affected. Elongation at break was used as an indicator of the degradation process because if the initial value of elongation at



**Figure 5** Degradation rate for carbonyl region along degradation process for ABS samples submitted to a) accelerated and b) natural weathering.



**Figure 6** Evolution butadiene amount along degradation time for samples submitted to a) accelerated and b) natural weathering.

break decreases 50%, the material is considered not suitable for any applications.

The mechanical properties of ABS films submitted to accelerated and natural weathering are shown in Figure 8(a,b), respectively. Although, degradation is a phenomenon that initially occurs at the material surface, it is known that the UV radiation leads to damages inside the sample and, consequently, there is loss in mechanical properties.<sup>16</sup>

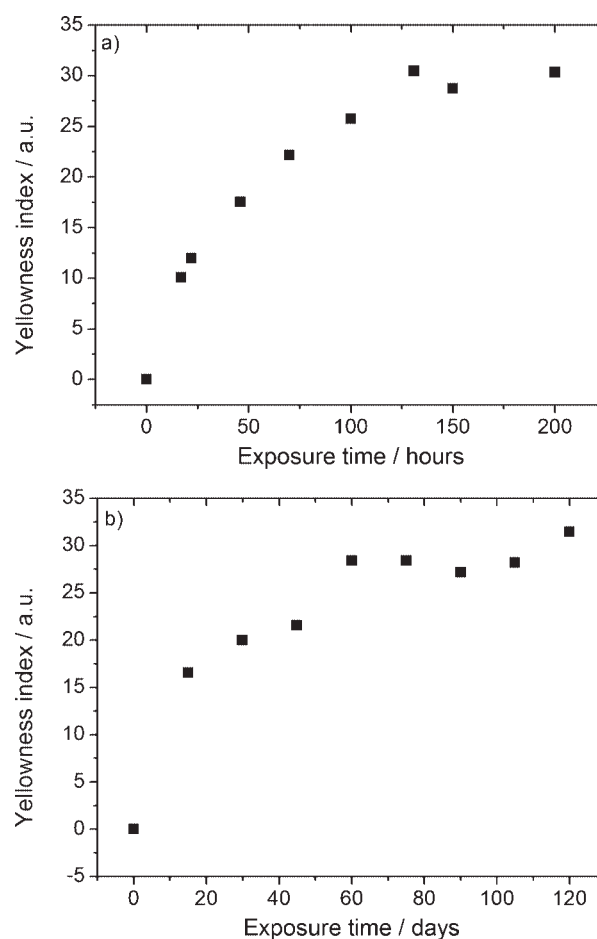
Whereas ABS films submitted to accelerated weathering lose approximately 60% of elongation at break after 22 h of exposure [Fig. 8(a)], samples exposed to natural weathering have a decrease of elongation at break of approximately 80% after 15 days. The drastic loss of deformation capability observed in the first exposure hours makes the copolymer mechanically fragile and inadequate for several applications. This behavior can be explained by the chemical change that occurs initially on the material surface, being the photodegradation of butadiene component the main reason for the loss in mechanical properties of ABS. According to Figure 6(a,b) the butadiene content after 22 h and 15 days

is approximately 17% and 9% of the original amount, respectively.

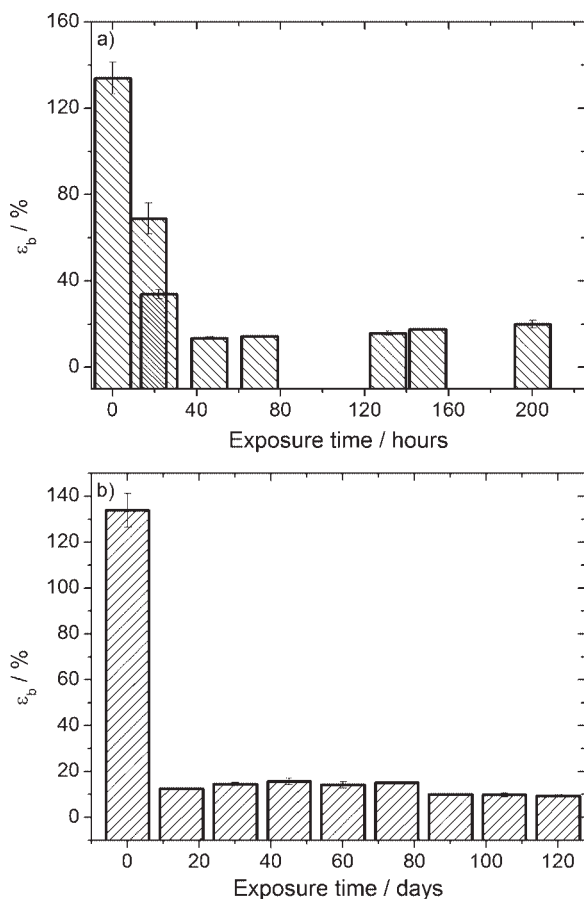
The results obtained show that the ABS oxidation surface has an important influence in mechanical performance. The high decrease of elongation at break during both types of weathering can be explained also taking into account that degradation process induces surface fragility, which results in cracks formation and propagation within the polymer.

Even though the decrease of butadiene content is the main reason for mechanical loss, the rheological characterization was performed to obtain more details on the degradation process. The evolution of complex viscosity and storage modulus in a frequency range of the samples submitted to accelerated and natural weathering is depicted in Figures 9 and 10.

The results show a different evolution of the rheological parameters along the time. While for samples submitted to accelerated weathering no significant changes can be noticed [Fig. 9(a,b)] until 150 h, samples exposed to natural conditions show a different behavior. A slightly decrease of complex viscosity



**Figure 7** Influence of the accelerated (a) and natural (b) weathering in the yellow index of the ABS films.



**Figure 8** Elongation at break for ABS samples submitted to accelerated (a) and natural (b) weathering.

and elastic modulus from 0 to 15 days can be observed and a significant decrease was observed until 120 days. These results suggest that chain scission may take place during natural exposure. The rheological behavior is in agreement with mechanical tests, the higher loss in mechanical properties of the samples exposed to natural conditions can also be associated with a decrease in rheological properties. Thus, besides the decrease of the butadiene content, the loss in mechanical properties can also be due to chain scission and cracks formation, and its propagation along the thickness.

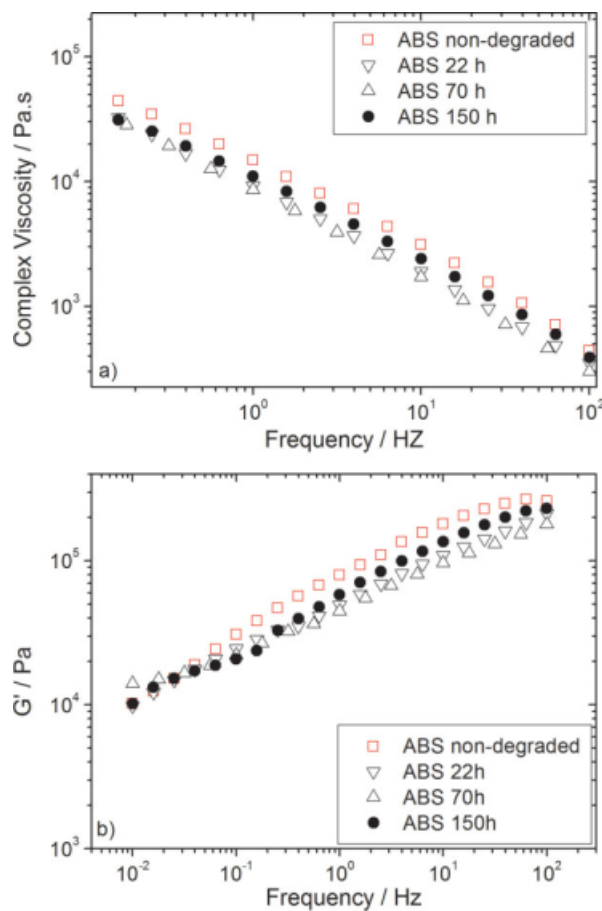
The difference between accelerated and natural weathering results can be explained taking into account the different parameters present in each type of aging. As it can be seen in Tables II and III the main difference are temperature, rain amount, relative humidity, and UV exposure time. On one hand, the radiation intensity and, consequently, temperature is higher under accelerated conditions, which enhance the formation rate of degradation products. On the other hand, natural weathering seems to be extremely influenced by rain amount, relative humidity, and several other factors that are not present in accelerated equipment, such as,

atmospheric pollutants (ozone, smoke, sulfur dioxide, nitrogen oxides, and heavy metals), and mechanical stress (related to rain, hail, and wind).

### Thermal properties

The ABS is an amorphous copolymer that presents two characteristic glass transition ( $T_g$ ) temperatures: one at  $-58.1^\circ\text{C}$  related to the butadiene component and another at  $112.2^\circ\text{C}$  corresponding to acrylonitrile-styrene macrophase.<sup>38</sup> Although, larger modifications were expected on  $T_g$  corresponding to butadiene component as it is the major responsible for ABS photodegradation. Due to equipment limitations it was only possible to study the influence of the chemical changes that occurred along degradation on  $T_g$  related to the acrylonitrile-styrene macrophase.

Table IV shows the values of  $T_g$  obtained for samples exposed to both types of weathering. While in samples exposed to accelerated weathering no changes were detected after 150 h, samples exposed to natural weathering during 120 days present a



**Figure 9** Elastic modulus and complex viscosity of samples exposed to accelerated weathering. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]

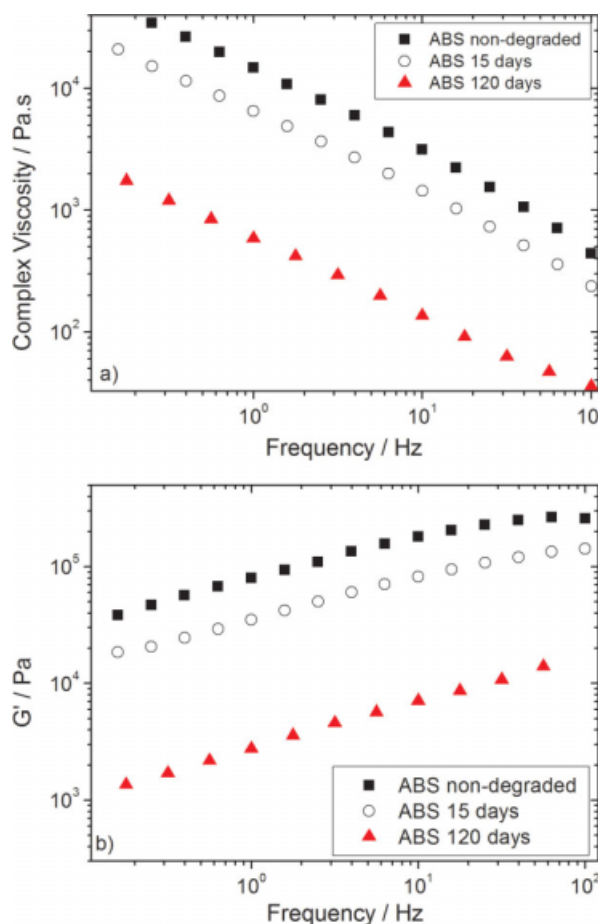


lower value of  $T_g$  of the acrylonitrile-styrene macro-phase. The decrease of the  $T_g$  is more pronounced for the samples submitted to natural weathering, which is in accordance with rheological measurements, where a decrease of complex viscosity and elastic modulus occurred. As before, this behavior can be explained by the formation of molecular chains of smaller dimensions as a consequence of chain scission along the degradation process.

## CONCLUSIONS

In this work, the combination of several characterization techniques allowed to follow the effect of ultraviolet radiation on photodegradation of ABS films when exposed to UV radiation.

Although molecular structure modifications occurs preferentially on material surface, the results evidence that these surface phenomenon during degradation process affects considerable optical, mechanical, and rheological properties.



**Figure 10** Elastic modulus and complex viscosity of samples exposed to natural weathering. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]

**TABLE IV**  
Glass Transition Temperature ( $^{\circ}\text{C}$ ) of Nonexposed Samples and Exposed to Accelerated and Natural Weathering, Respectively

Degradation	Samples	$T_g \pm \Delta T_g$ ( $^{\circ}\text{C}$ )
ABS nonexposed		$102.80 \pm 0.68$
Accelerated weathering	ABS 70 h	$102.71 \pm 0.33$
	ABS 150 h	$101.32 \pm 0.42$
Natural weathering	ABS 90 days	$102.30 \pm 0.09$
	ABS 120 days	$100.46 \pm 0.93$

The drastic decrease of elongation at break for ABS samples exposed to both type of weathering is not only due to the decrease of butadiene amount but as a consequence of fragile and brittle surface, which leads to cracks formation and propagation within the material.

While the results obtained for samples submitted to accelerated weathering conditions show higher formation rate of photodegradation products, during natural conditions chain scission is more pronounced.

The results obtained through this work provide a strong incentive to develop an improved stabilization system for ABS, which will be accomplished in our future work.

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