RSSI, Vol. 06, No. 01, Décembre 2016 07-11

Revue des Sciences et Sciences de l'Ingénieur

ISSN 2170-0737

Journal homepage :http://www.RSSI.lagh-univ.dz

Effect of Weathering on the Viscoelastic Behavior of LDPE Films

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Abstract—Low density polyethylene films commonly used for greenhouses covering have been aged in sub-Saharan region. Mechanical characterization with a tensile test machine has been performed on polyethylene samples picked up at regular interval thorough the total duration of the aging protocol (0 to 6 months). Young's modulus, elongation at break and tensile strength are the different greatnesses which have allowed following the incidence of ageing on the mechanical properties. The viscoelastic properties were carried out on a creep apparatus. It has been observed that the film deforms according to several steps. They correspond to different flow types; such as instantaneous deformation, viscoelastic and viscoplastic deformation. All are affected with aging time in reason of the micro-structural changes undergone by the material.

Index Terms— LDPE, Weathering, Tensile strength, Viscoelesticity, Young's modulus, Creep's modulus.

I. Introduction

Low density polyethylene (LDPE) films used for greenhouse covering is an old research subject which has not finished revealing all the reasons of the loss of the properties of use. It is well known that the structural damages undergone by the material are mainly due to the environmental interaction with the film. Conventional mechanical testing methods such as tensile test give a good insight on the extend of the structural damages which adversely affect the ultimate mechanical properties and the elastic character as well. However, a rheological approach making possible the measurement of the different viscoelastic properties would bring additional information making possible to connect the impact of the molecular and the morphological changes on the mechanical behavior. Creep experiment can be regarded as a very powerful method to reach some greatness such as the creep modulus, the creep rate (strain rate), the instantaneous, the viscoelastic and viscoplastic deformation. In the present work are presented the results obtained by tensile test and creep experiment on plastic films outdoor exposed in sub-Saharan region. It is well known that the most deleterious region of solar light is that of the UV portion [1, 2]. On the light of the results obtained it has been tried to connect the effect of the structural changes undergone by the material due to ageing with the variation of the different mechanical properties.

II. Material and methods

Material

The raw material is a Low Density Polyethylene, LDPE, produced by the SABIC society under the trade name (2100 TN00 W) of Saudi Arabia. It has been shaped into films via a blow extrusion process performed by the Sofiplast Company of Sétif. The tichkness of the finshed product is about 164 µm, with a semi-crystalline structure [3] (i.e constituted with a crystalline and an amorphous phase). The tensile test specimens are cut in the machine direction for both unexposed and aged films. The total length of the tensile test specimen is 70 mm with a gauge length of 40mm and a width of 10mm. The creep test specimen is 110 mm length with a gauge length of 80 mm and a width of 10 mm.

Exposure

Exposure has been performed at Laghouat (33 ° 48'N 2 ° 52'E) (Algeria). The films were fixed on a white metal support to avoid overheating in the contact zone. The support is inclined by 33 ° and oriented to south to get a maximum duration of solar irradiation (NF51-165) (Fig.1). The duration of the aging protocol is about six months (December to June) limit at which the film starts to become too brittle to be handled. Every month a sampling is performed.

Fig.1: Natural irradiation device.

Mechanical tests

Tensile tests have been performed on a traction machine (Zwik / Rowell) with a cell force of 1kN. To reach the ultimate properties (i.e. Stress and strain at break; Yield stress) the crosshead speed was fixed to 200 mm/mn. For the modulus it was fixed to 2mm/mn [**NFT 54-102**].

The creep tests were performed in the laboratory at 15 ± 2 °C (temperature well above Tg ≈ - 110 °C). The creep apparatus allows monitoring the progressive deformation of the test pieces to which a load of 1 500 g has been hung (Fig.2). A time of 8hours for the creep have been applied for all the samples. These time and load have been optimized to make possible to test the most aged samples (6 months). The total deformation is directly indicated by the apparatus. The relative deformation of the test pieces is measured by means of ink marks drawn on the surface of the sample and recorded by means of a camera which follows continuously their respective progressive motion.

The pictures are then processed by means of the Matlab software.

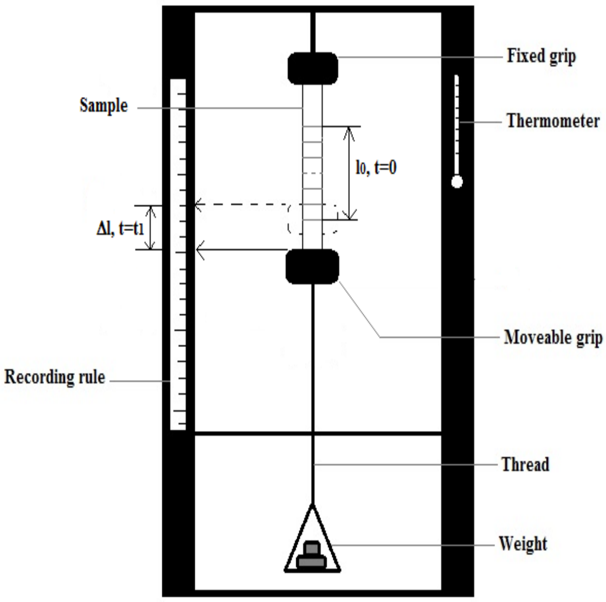

Fig.2: Creep tester.

III. Results and Discussions

Tensile tests

The stress-strain curves of the unaged and aged films are reported in Fig.3. The Young's modulus E, the Yield stress σ_y the stress and the strain at break σ_b, ε_b, are listed in Table 1. With ageing the ultimate mechanical properties decrease drastically. The Young's modulus relative to the low elastic deformation of the film increases progressively with exposure time. Its progression is not monotonous since it is very slow during the two first months to suddenly increase the fourth month to about 8 %. Then the progression lowers and reaches only 2.5 %. However, the overall progression of the modulus is around 10 to 11%. This overall progression shows the progressive stiffening of the material.

The decrease of the ultimate properties which leads to a rapid lowering of the surface of the *((σ-ε)* curves reveals the progressive transition of the mechanical mode of fracture of the films which pass from a ductile to a very brittle mode of fracture.

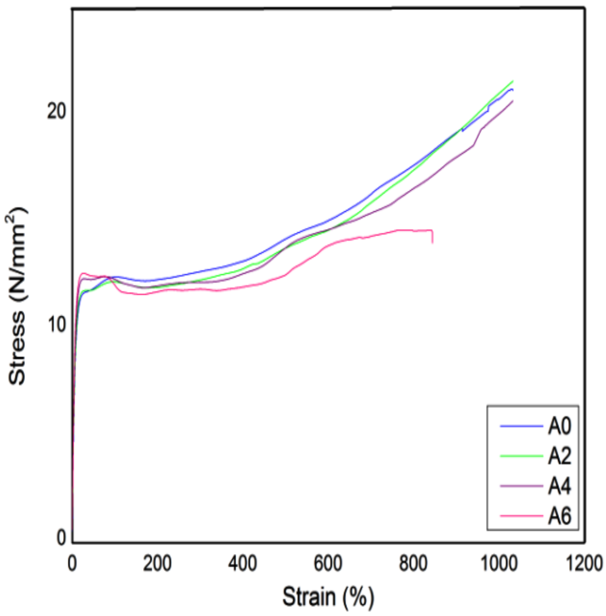

Fig.3: ((σ-ε) curves of aged samples (0; 2; 4; 6 months).

Table 1: Mechanicals Properties of Different Samples (0; 2; 4; 6 months).

	A0	A2	A4	A6
E (MPa)	198	201	217	221
(MPa)	11.76	11.82	12.38	12.63
(MPa)	20.53	19.7	19.74	11.55
ε_b (%)	1019.21	948.25	986.79	632.88

The σ_y, σ_b and ε_b , which are representative to the capability of the material to support the deformation, progress irregularly with ageing. The initial strain at break value is 1019 %, with a corresponding stress of 20 MPa. These two values denote the highly ductile character of the unaged material. Elsewhere, by other of us it has been shown that a deformation rate, up to 600 % can be reached by a same kind of PE films [3], however this value remains relatively lower compared to that of the material used in this work. If the general trend of the mechanical properties is to decrease with ageing time it can be still observed a slight strengthening after 4 months; then they definitely diminish.

The overall decrease rate of the elongation at break is 400% and 43% for the stress at break. The sudden drop of these two greatnesses after four months is very indicative to the extent of the damages caused to the molecular and the morphological structure. Among them one can quote oxidation and chain scissions (reduction of the chain length) responsible to the reduction of the average molecular weight leading in most of the cases to the increase of the crystallinity via a chemo-crystallization process [3]. Stiffening revealed by the Young's modulus is due in one hand to crosslinking reactions via the consummation of the vinylidenes and in another hand to the increase of crystallinity. Strengthening observed at the fourth month can also be assigned to crosslinking reactions while the increase of crystallinity would per contra affect it adversely. As such, the sudden drop of all the mechanical properties are indicative of the strong microstructural changes induced by UV exposure. It is generally estimated that an overall loss of the strain at break of 50% can be considered as an end of life criterion [1, 3].

Creep tests

The creep curves of the film for different ageing stages (0, 2, 4, 6 months) are reported on Fig.4.

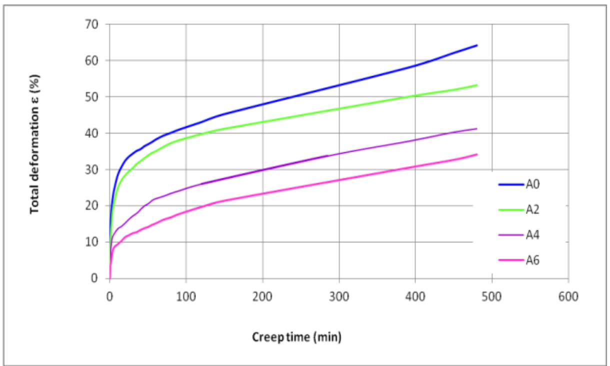

Fig.4: Creep curves of PE film for different ageing stages (0; 2; 4; 6 months), under a load of 1500g (T=16°C).

The viscoelastic character of the film contributes significantly to the creep response. For the unaged film (A0) the total deformation reaches almost 60% divided in two different portions, 18% for the instantaneous deformation; 42% for the delayed deformation. The delayed deformation starts quickly just after the instantaneous deformation and then increase more progressively until the end of this phase. Also, it is currently admitted that the first part of this phase corresponds to the viscoleastic deformation while the second part is relative to the viscoplastic deformation [4]. The evolution of these different types of deformation is reported in table 2. It can be observed that the instantaneous ($\varepsilon_{elastic}$) and the visoelastic ($\varepsilon_{viscoelastic}$) deformation decrease with exposure time. Although, the viscoplastic deformation ($\varepsilon_{viscoplastic}$) decreases overall, it still progresses irregularly.

Table 2: Elastic, viscoelastic and viscoplastic deformation as function of ageing time.

	A0	A2	A4	A6
$\varepsilon_{elastic}$ (%)	18.073	16.81	9.32	8.46
$\varepsilon_{viscoelastic}$ (%)	13.607	12.08	9.47	5.85
$\varepsilon_{viscoplastic}$ (%)	27.91	18.74	21.23	20.86

To highlight the different parts of the delayed deformation, the strain rate has been plotted versus the creep time (Fig.5). A cursory observation of the $(\dot{\varepsilon} - t)$ curves let appears different domains for the creep rate which results in a variation of the slop of the concerned curve segments.

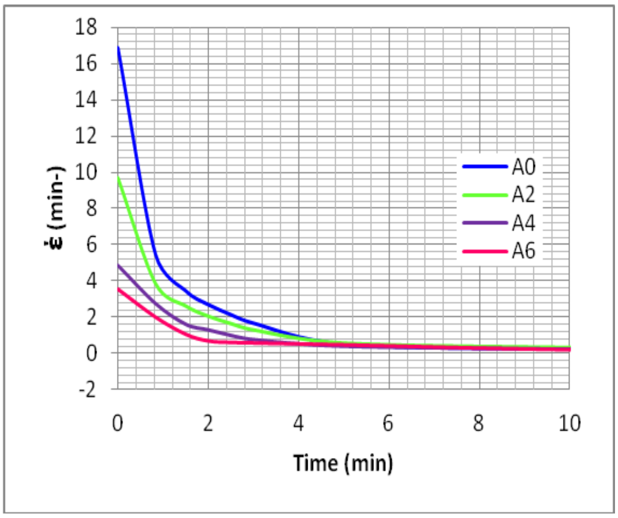

Fig.5: Creep rates as a function of time for different aging states (A0, A6) for an applied load of 1500g.

It appears that it progresses according to two distinguishable phases. For all the ageing stages the first phase is rapid in the beginning and then decreases progressively until to vanish (primary creep). Above this limit of time the second phase starts where the strain rate reaches and remains zero (secondary creep). It can be observed that with ageing secondary creep begins earlier.

In the first phase the creep rate is very rapid for the unaged sample and decreases progressively for the more aged ones. The first part of this phase is a linear slope which lowers rapidly. The second part is characterized by a slow-down of the creep rate describing a parabolic slope.

In other words one can say that ageing lowers the total deformation of the film and its rate as well. The shape of the curves total strain vs creep time and strain rate vs creep time, give a good insight on the structural changes undergone by the material. In fact, chain scissions which are predominant for an advanced stage of ageing reduce the average molecular weight. This has an adverse impact on the viscoelactic properties. The number of tie molecules linking the crystalline lamellae becoming lower and shorter, the material tends to loss its viscous character.

Others have demonstrated that for the same type of material an increasing concentration of short chain branching decreases the creep rate [5].

The ratio of the stress on the strain allows finding the creep modulus. Also, its graphical representation for the different ageing stages makes possible the visualization of this greatness variation with the creep time (Fig.6). It appears that in all the cases the modulus deceases with the creep time.

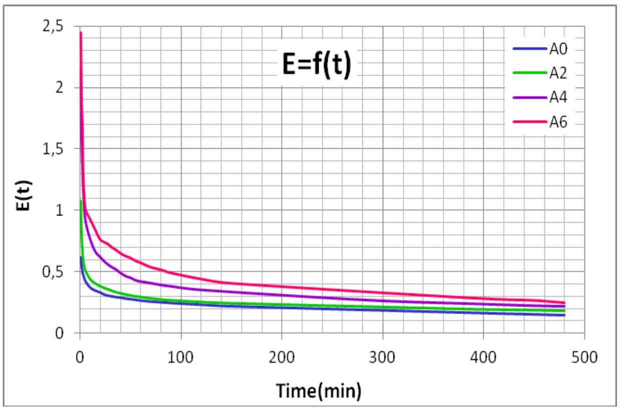

Fig.6: Creep's modulus versus time for different states of weathering.

They all progress in the same way and they appear distinctly one above the other. The order of the curves superposition goes from the less aged film to the more aged one. This clearly indicates the stiffening of the material with ageing. These results consolidate what have been found by tensile testing were the Young's modulus, the stress and strain at break (σ_b et ε_b) all argue in favor of a stiffening of the material accompanied by a loss of its ductility [3].

IV. Conclusion

The study of long-term behavior of LDPE, which remains very underdeveloped, has been conducted in order to contribute in the understanding of the viscoelastic behavior of LDPE films used for greenhouse covering. The tests have been performed at room temperature (which is well above Tg) and near to the temperature of use. The results revealed that with ageing the material stiffens and loss its ductility. Also, it passes from a ductile mode of fracture to a brittle one. The creep study appears as a very powerful means to show the loss of the viscous character of the material acquired with ageing.

References

[1] S.F Chabira, M.Sebaa, C.G'sell "Influence of climatic ageing on the mechanical properties and the microstructure of low density polyethylene" journal of APS November 2008.

[2] A.lounas, M.Hadjaj, M.Dakmouch, M.Saidi, M.Sebaa "natural aging of polyethylene film used as cover agricultural emissions in the region of Ouargla" Vol.4 No. 2, November 2012.

[3] S.F.Chabira, M.Sebaa, R.Huchon, B.Dejose "The changing anisotropy character of weathered low-density polyethylene films recognized by quasi-static and ultrasonic mechanical testing", SD volume91, issue8, pages 1887-1895,August 2006.

[4] Y.B. Unigovsky, A.L. Bobovitch, E.M.Gutan, D.Moginalsky "Effect of processing and density on morphology and creep behavior of LLDPE" Polymer enginering and science, 2011.

[5] G'Sell C., Dahoun A., Evolution of microstructure in semi-crystalline polymers under large plastic deformation, Materials Science and Engineering, A175, (1994), 183-199