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ORIGINAL ARTICLE

Durability of mono-layer versus tri-layers LDPE films used as greenhouse cover: Comparative study

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Abstract The mono-layer low density polyethylene LDPE films are often used as a cover for the greenhouses. Recently, there are few attempts for utilizing the tri-layers LDPE films as substitute for mono-layer film. The degradation and fracture behaviour with regard to abrasion of that new generation of polymeric greenhouse covers presenting a sandwich structure made of three layers of LDPE have been studied and compared with that obtained for monolayer film often used in North Africa for plasticulture devices. Surface analysis and mechanical properties have been analysed on samples having undergone natural conditions in the north of Africa (Algeria). The results revealed that the degradation performance of these new tri-layers films is found to be quietly better than that of the mono-layer film, with regard to the mechanical and surface energy properties. The lifespan of these films under natural conditions in the north of Algeria is estimated to be 10 and 5 months for the tri-layer and mono-layer films, respectively.

1. Introduction

There is an increasing demand on the use of greenhouses due to their impact on agriculture. The main component in the greenhouse is its cover film. Therefore, studying the performance and durability of such structure component material under different ageing conditions leads to optimising the process parameters of these films and its structure. Indeed, this will improve the performance efficiency and lifetime of the films and consequently its impact on the economy. Usually the cover film is produced as a mono-layer of low density polyethylene. There are three main categories of conditions (factors) that affect the physical and mechanical properties of polyethylene films used as greenhouse covers. These can be grouped as: (a) product manufacturing and process specifications, (b) greenhouse external climate conditions and (c) greenhouse microclimate (internal) conditions. Dilara and Briassoulis (1998) have shown that manufacturing process

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parameters of plastics that affect the mechanical properties of the film include temperature of the melt, die parameters, blow-up ratio, drawn ratio and frost-line height and cooling conditions.

Gruenwald (1992) and Vishu (1995) have shown that Molecular orientation during film blowing influences tensile properties; higher in the direction of the covalently bonded carbon–carbon chain than in the transverse direction that is dominated by weaker van der Waals bonds. Climatic conditions, such as solar irradiation, temperature, humidity, rain, wind loads and environmental pollution influence ageing and mechanical properties of low density polyethylene (LDPE) greenhouse covers. Khan and Hamid (1984) have reported that the Ultra Violet UV radiation (290–400 nm) is the high energy part of the solar spectrum, and can be absorbed by the polyethylene. This leads to bond cleavage and polymerization, causing photo-degradation (oxidation) and thus mechanical degradation. Cyclic temperatures, high when in contact with metal greenhouse frames during hot and sunny days, but low at night might increase degradation (Dilara and Briassoulis, 1998). Dilara and Briassoulis (2000) illustrated that other external weathering factors include photo-induced degradation, thermal and mechanical stresses, ultrasonics and vibration, and hydrolysis. The third group influencing mechanical properties of greenhouse polyethylene is microclimate conditions, such as internal humidity and temperature, biological activity and agrochemicals. A combination of such harmful factors can eventually alter the properties of the polymer and affect adversely its mechanical and optical properties. It should be considered that various testing methods of researchers and approaches possibly introduce variations in data. The available tests for mechanical and physical properties of plastics used as greenhouse covering materials are mainly for general purpose and are not in European Union standards (Dilara and Briassoulis, 1998). There is little information on the type of tests recommended for greenhouse covers. Mechanical degradation of greenhouse polyethylene can be of several forms that include rupture, tearing, shearing, penetration, impact, or excessive elongation. Much effort is needed to standardize mechanical tests for greenhouse covers, especially to account for degradation due to ageing.

Hassini et al. (2002) have studied the effect of simulated sand wind for a duration of four hours on a polyethylene film. Briassoulis and Schettini (2002) have utilized strain gauges as a mean for measuring the deformation in the film subjected to mechanical stresses. Salem (2001) examined the effect of UV radiation on the mechanical properties of LDPE films containing black carbon titanium oxide. The UV radiation has changed the elongation and shear stress of the samples. Pacini (2001) emphasized the importance of keeping the plastic greenhouse cover in good conditions and examined covers' properties including total permeability to solar radiation, mechanical properties, shelf life, effect of climate, and the homogeneity of film thickness and width. Shen et al. (2000) developed a simulation model to calculate the distribution of the molecular structure at the inner surface of the greenhouse cover and to calculate the yield and elasticity. This was to predict the maximum ice and storm loads on the greenhouse structure. Nijskens et al. (1990) discussed the effect of ageing on the mechanical properties (maximum values of the tensile force, elongation, and fracture force) of greenhouse polyethylene covers. Mourad et al. (Mourad, 2010; Mourad et al., 2005, 2009a, b; Fouad et al., 2008) have studied the mechanical, thermal and chemical stabilities of different grads of polyethylene under different ageing conditions.

The final design of a greenhouse should fulfil a balance between three following important issues: (a) the overall structural design of the greenhouse and the individual structural components characteristics, (b) the inherent mechanical and physical properties which determine the structural and the functional behaviour of the cladding materials, and (c) the specific sensitivity to light and temperature of the crop to be grown in the greenhouse along with other agronomic requirements (Briassoulis et al., 1997). Therefore, the evaluation of the mechanical properties of the greenhouse covering material combined with their physical properties under the environmental factors involved is critical in assessing their quality and in determining the relevant requirements concerning the structural design and construction details of a greenhouse.

Generally, and based on the previous literature, UV radiation, external pressure, humidity, and high temperature shortened the shelf life of the covers. There is little information on the influence of arid environmental conditions of ageing on the behaviour of greenhouse polyethylene covering. Further the previous studies have been conducted mainly on mono-layer polyethylene films in moderate climates considering different amounts of additives to improve their performance. Few authors were interested in long term behaviour of polyethylene films [e.g., 7]. Recently tri-layer LDPE films have been considered by Dehbi et al. (Youssef et al. 2008a, b, Dehbi et al., 2010). They have studied the effect of different arid conditions under long term (up to 5400 h) of natural and artificial ageing on the behaviour of LDPE films as a greenhouse cover. The films were produced in Algeria by Agrofilm Company using a co-extrusion process with some additives such as anti UV and, anti oxidizing agents. In general the greenhouse covering should be durable, strong enough to resist loads due to snow, wind, crops and installation and with acceptable lifespan. The mechanical properties of greenhouse coverings are very important in relation to their mechanical behaviour under various loading conditions as well as to the overall structural behaviour of the greenhouse. Therefore, the focal point of this work is to conduct a comparative study between both mono-layer and tri-layers films in terms of the degradation in their mechanical performance under different natural ageing conditions.

2. Materials and methods

Two different films, produced and supplied by Agrofilm SA (Sétif–Algeria), were used in this study. The first is extruded mono-layer low density polyethylene LDPE film (before extrusion) that has density of 0.923 g/cm^3 and the weight average molecular weight is in the range of 90,000–120,000. The melt flow index MFI of the raw LDPE is 0.33 g/10 min and the MFI with stabiliser is 10 g/10 min, with a thickness of 180 μm. The second is three co-extruded layers LDPE film with the same thickness of mono-layer film (180 μm) and with the following proportions in the layers: 1/4, 1/2, 1/4. The real film composition is not known (kept confidential by the supplier) but it is established that the two exterior layers have different additives than those added into the medium layer. The various used additives are anti-UV/UV stabilizer, antioxidant/anti-O_2, Nickel stabilizer, plasticizer, etc. The density of LDPE before extrusion is 0.923 g/cm^3. The initial colour of the film is milky yellow.

Two greenhouses of 32 m length, 8 m width and 3.5 m hight have been built specially for studying the effect of the natural ageing on the performance of the LDPE cover. Samples have been taken every month over a duration time of seven months. To ensure the data reproducibility, large square samples of 30-cm sides were cut, at each ageing period, from the cover from which sufficient test samples were prepared to conduct the experiments required. The climatic conditions undergone by the roof are displayed in Table 1. The climatic condition was classical and standard in North Algeria.

The tensile tests were performed using a universal testing machine (Instron model 4301) with a load cell of 5 kN. All tests have been performed at room temperature with a cross head speed of 50 mm/min and displacement controlled conditions.

3. Results and discussion

The tensile tests were conducted on unaged/virgin and natural ageing samples (up to a period of 7 months) to obtain a quantitative estimation of the effects of natural ageing on the mechanical performances of the mono-layer and tri-layers films. The load was applied on the specimen in a direction parallel to the average molecular orientation obtained during the film processing. The engineering stress as a function of the engineering strain has been recorded for each film. The tests were conducted to determine the tensile properties (modulus of elasticity E, fracture stress, and elongation at break) of the material.

Fig. 1 shows the stress–strain diagrams for unaged and naturally aged mono-layer films and Fig. 2 presents the curves for tri-layers films. The curve for unaged film is included in Figs. 1 and 2 to illustrate the effect of different ageing periods on the properties of both types of films (mono-layer and tri-layers) if compared with that for the unaged films.

The tensile curve of mono-layer film (Fig. 1) can be divided into four distinctive regions. In the first region the stress varies with the strain following a linear relationship. In this region Hook's law may be applicable and valid up to the proportional limit at which the curve deviates the linearity and the second non linear variation region starts. In the third and forth regions the stress varies almost linearly with the strain, however, the slope is less in the fourth region). All samples exhibit the same tensile behaviour trend regardless the ageing conditions.

For the tri-layers film, the stress–strain diagram increases almost linearly in the 1st (initial) region. Then the curve becomes nonlinear (in the 2nd region) with decreasing slope up to a point of zero gradient. Then the stress drops little to a constant drawing stress or becomes almost with zero gradient (in the 3rd region) up to a certain engineering strain prior to the work hardening starts and the stress increases almost linearly with the strain in the 4th region up to fracture takes place. As for mono-layer, all samples exhibit the same tensile behaviour trend regardless the ageing conditions.

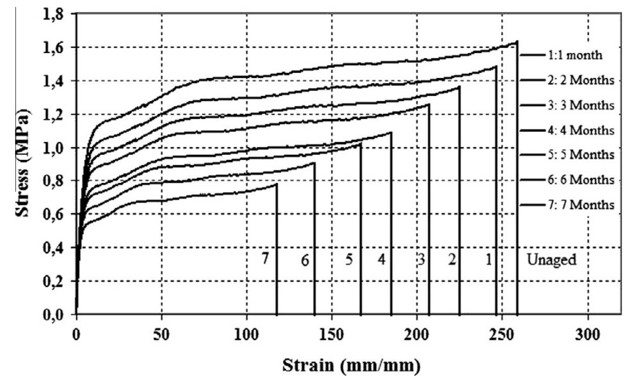

Figure 1 Stress–strain curves for unaged and aged mono-layer LDPE films.

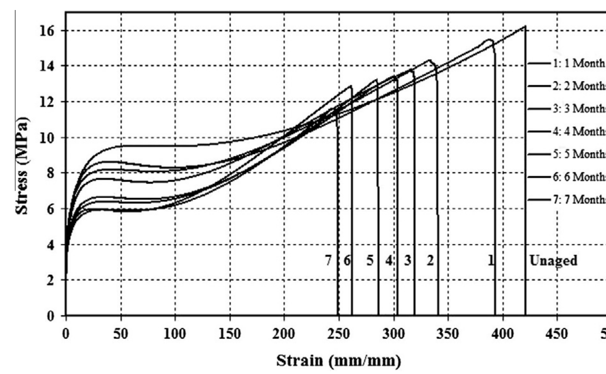

Figure 2 Stress–strain curves for unaged and aged tri-layers LDPE films.

Figs. 1 and 2 show that, the unaged film has a higher level of stress than aged films. The curves reflect the deteriorative effect of ageing on both types of LDPE films. The harshest effect (in terms of the reduction in the yield and fracture strengths and elongation at brake) is after seven months of ageing. The ageing also increases the stiffness of the films on the expense of ductility.

For the purpose of comparison between the deteriorative effect of ageing on the mono-layer and tri-layers films, each two corresponding curves are plotted together at different ageing periods as shown in Figs. 3a–h. The levels of the stresses at the same ageing period for tri-layers films are quite higher than those for the mono-layer. Further, the tri-layers films show a quite higher level of ductility. On contrast, the stiffness (elastic modulus) increases with ageing which reflects a measure for degradation in the flexibility rather improvement in the

Table 1 The average temperature and average moisture during ageing.

Month	1 (March)	2 (April)	3 (May)	4 (June)	5 (July)	6 (August)	7 (September)
T (°C)	32.8	33.6	37.6	41.6	44.2	43.6	41.1
H (%)	76.2	69.3	62.5	60.58	58.7	59	63.7

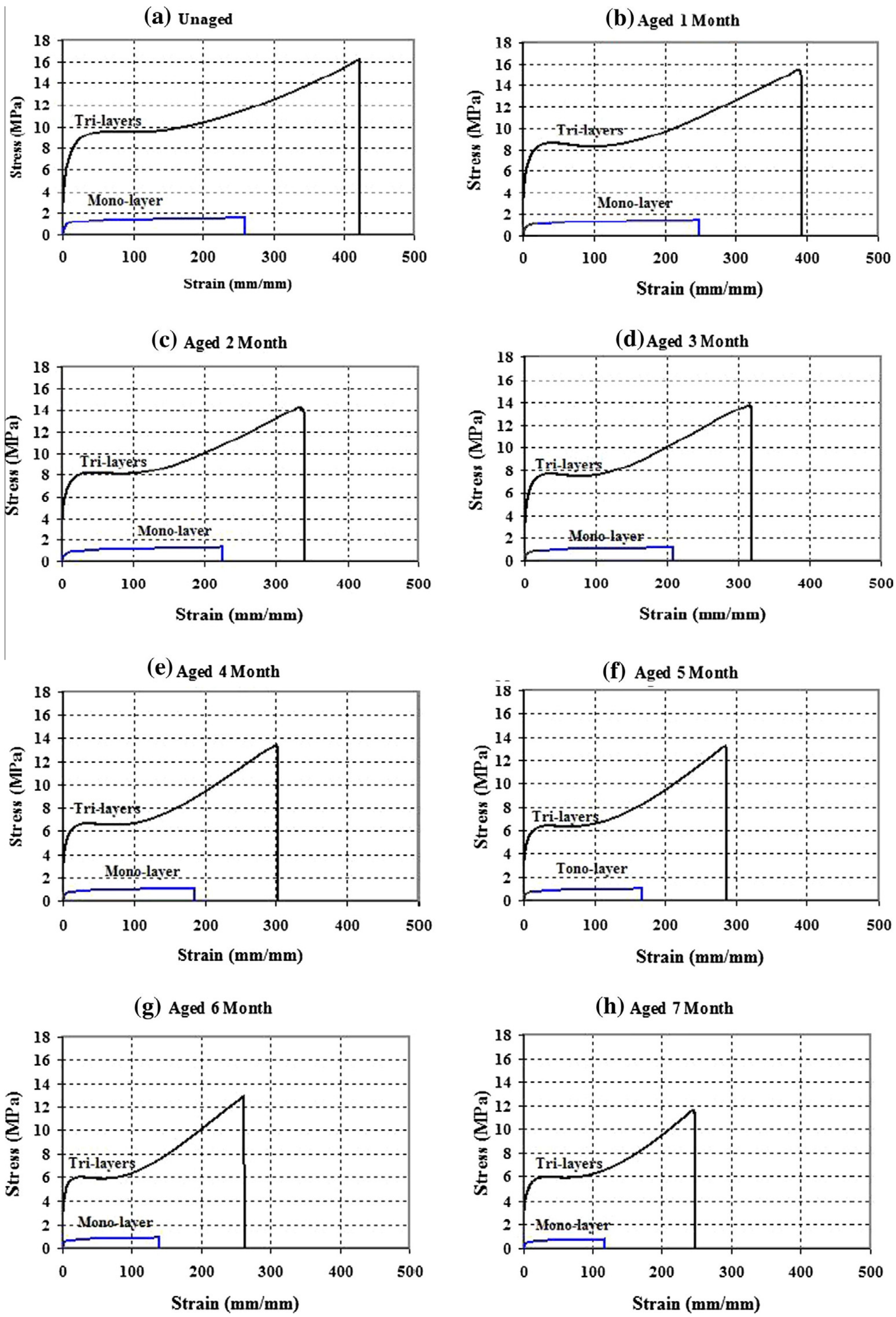

Figure 3 Comparison between stress–strain diagrams of mono-layer and tri-layers films at different ageing periods.

stiffness. This will be discussed from a quantitative point of view later in the following sections.

Fig. 4 shows the variation of the fracture stress/the stress at break with the ageing time. The fracture stress reduces with ageing time. The trend of variation resembles that for the yield strength. The maximum achieved fracture stress for unaged tri-layers and mono-layer films are 16.2 and 1.63 MPa, respectively. This means that the fracture stress of unaged tri-layers film is 9.9 greater than that of mono-layer film for the same thickness (180 μm). The corresponding values after 7 months of ageing are 12.51 and 0.5. In sense the fracture stress of tri-layers is 25 times that of mono-layer after exposure of 7 month of natural ageing. The tri-layer lost about 22% of its value for the unaged film and the mono-layer lost 69%. The results again demonstrate that the fracture stress of the tri-layers film is essentially higher than that for the mono-layer one.

Fig. 5 demonstrates the impact of ageing on the strain at break. The trend of variation is similar to that of the fracture

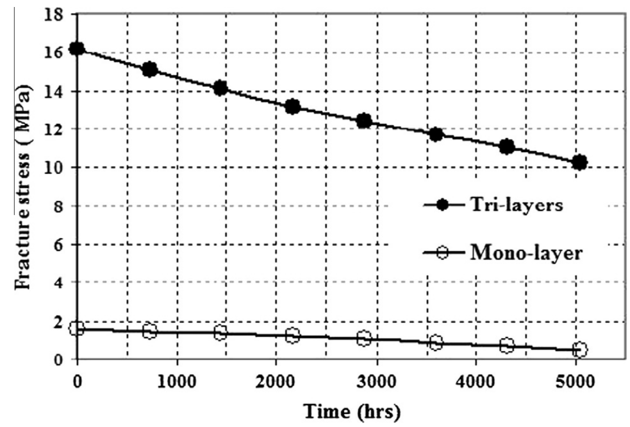

Figure 4 Variation of fracture stress with ageing time.

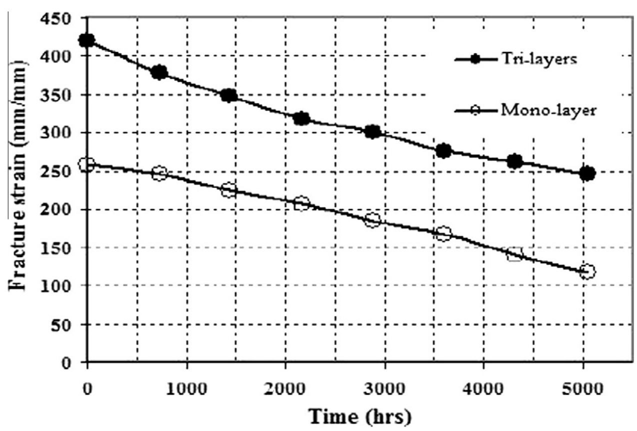

Figure 5 Variation of the fracture strain with ageing time.

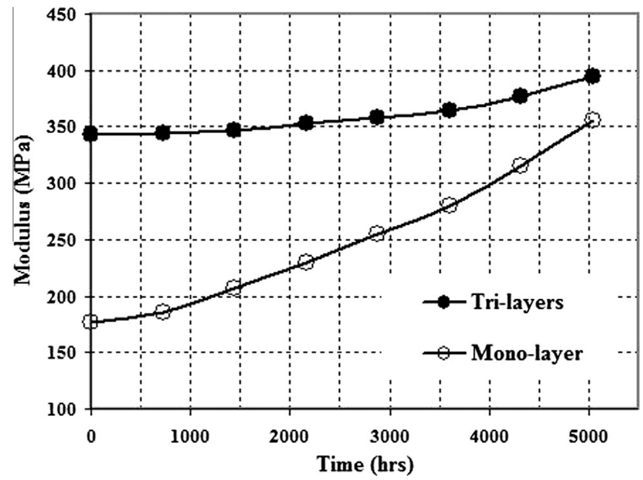

Figure 6 Variation of modulus of elasticity E and with ageing time.

stress (Figs. 4) for both types of films. The unaged tri-layer film exhibits a remarkable ductility (420% elongation). Though the mono-layer exhibits also a ductile behaviour (259% elongation), yet the ductility of tri-layer is 1.63 greater than that of mono-layer. This ductile behaviour diminishes with ageing. The percent elongation of both types of films is 258% and 118%, respectively, at the end of the total ageing period. In sense, the tri-layers film% elongation to the mono-layer % elongation ratio is 2.18. It is worth noting here that the percent elongation of tri-layers film after 7 months of ageing is approximately of the same order of that of the unaged mono-layer film which again reflects the notable ductility of tri-layers with respect to that of the mono-layer even after ageing.

As all curves of mono-layer and tri-layers films exhibit almost an initial linear relationship, the modulus of elasticity E is determined as the slope of the first portion of the tensile curve. The variation of the modulus of elasticity E with ageing time for the tri-layers and mono-layer films is given in Fig. 6. The value of the modulus of elasticity was found to be 343 and 177 MPa for unaged tri-layers film and mono-layer film, respectively. The values increase with ageing time. The elasticity modulus is 445 and 355 MPa after the 7 months of ageing. The modulus for the aged mono-layer is of the same order of that for the unaged tri-layers after the total ageing duration. The value for the tri-layers increased by 23% of that of the unaged film and the corresponding value for the mono-layer is 50%. Furthermore, the curves compromise for two linear regions. The rate of degradation in the flexibility in the second region is higher. The second region commences after 3000 h of ageing for the tri-layers film and 700 h only for mono-layer film, respectively. This reflects the severe impact of ageing on the flexibility specially for the mono-layer as the increase in the stiffness is undesirable property.

Studying the impact of the ageing on the performance of the greenhouse cover is an important issue. However, it is with essential importance to set a criterion to evaluate its lifetime. The life time of the cover should, of course, be based on the loss of its physicochemical and mechanical properties. Briassoulis (2006) has shown that if the greenhouse cover film loses 50% of one of its original/initial properties (i.e., for unaged film), it becomes then unusable and is said to be failed. For such purpose and to compare between the lifetime of mono-layer and tri-layers LDPE films, their mechanical properties have been normalized with the corresponding value of the unaged film and plotted in Figs. 7a and b. Fig. 7a demonstrates that the aged mono-layer film loses 50% of its mechanical properties comparing with that of unaged film after exposure time between 3200 h (as lower shelf) and 4200 h (as upper shelf). In average, the lifetime is around 3700 h (5 months) which is just sufficient for only one agricultural turn. It should be noted that the 50% increase in the modulus of elasticity (which is undesirable increase) corresponds to 50% loss in the flexibility.

Fig. 7b presents the evolution of the normalized mechanical properties with ageing exposure time for tri-layers films. Unlike the mono-layer LDPE cover, the 7 months exposure time to the natural conditions were insufficient to cause a loss of 50% in any of the mechanical properties. The work is currently in progress to cover this period. Therefore, a fitting curve technique is used here to predict the ageing time at which the 50% loss in the property occurs. This technique has been used satisfactorily in previous work by Dehbi et al. (2010). Some fitting equations have been tried to fit the experimental data. Ultimately, the following equation has been found to be the best one for fitting the experimental results and extrapolating the data beyond the exposure time at which the 50% loss in the property takes place.

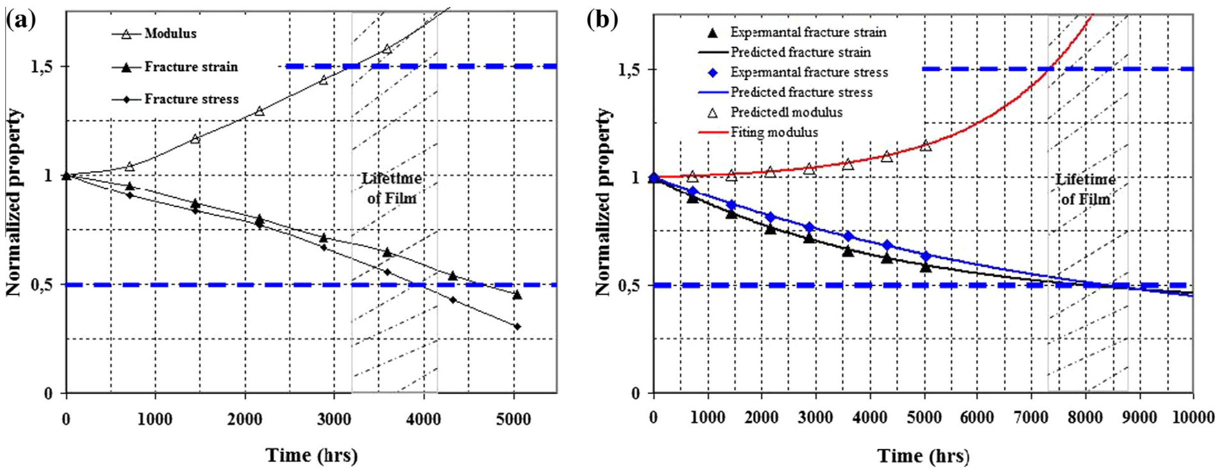

Figure 7 Variation of the normalized mechanical properties with ageing time.

$$y(t) = a + b\exp(-t/\tau) \quad (1)$$

Where $y(t)$ is the property/parameter as a function of time t, a and b are the constants of normalization, τ is the time beyond which the rate of degradation becomes faster.

Fig. 7b shows that all normalized properties degrade with time. Fig. 7b shows also that the 50% loss of initial value of various parameters occurs between 7200 h (lower shelf value) and 8800 h. In average all normalized properties lose 50% of their initial values at ≈8000 h (≈10 months). It means the tri-layers film can be used as a cover for almost two agriculture turns (cultivation of two crops). Further the tri-layers film can serve efficiently for a period of time double than that for mono-layer film (3700 h/5 months). Generally, it can be concluded that the tri-layers film has a longer lifespan and more resistive to the normal ageing climatic conditions of north of Africa.

It has been confirmed from the company (Agrofilm) that the various important additives are anti-UV, anti-O_2, Nickel stabilizer, plasticizer, yellow colour additives. These various additives added during the film production and to protect LDPE against irradiation are able to diffuse from the bulk to the surface. In fact, yellow additives are also not stabilized and can migrate easily to the film surface. This is the main change that occurs for the material during ageing. Later, as the anti-UV additives are practically disappeared, the UV radiation and the associated oxidization reaction take place and accelerate and dominate the ageing process at the surface and through the thickness (Scoponi et al., 2000).

It is well known that there is a formation of radicals on the LDPE film surface due to the light and/or heat effects. These radicals lead to three different reactions that are responsible for degradation. These are reactions of reticulation of chains, reactions with oxygen in air and reactions of scission of chains. These three types of reactions can coexist. The prevalence of a mode of degradation with respect to another depends on the nature and the stability of the radical formed in the course of the reaction of the photo-oxidation (Dilara and Briassoulis, 1998). In the case of LDPE the tertiary carbons (20–40 carbon atoms/1000) can lead to stable radicals. They are likely to react with other radicals and to lead to reticulations of the chains. The secondary carbons lead to very reactive radicals and cause reactions of chains scission (Hanafi and Paposolomontos, 1999). Tidjani (2000) showed clearly the degradation model of polyethylene. This result supports the possible formation of vinyls by an intramolecular decomposition on sec-hydroperoxides. Vinyls can also be produced from the Norrish type II reaction on ketones. UV fractions of the solar radiation are absorbed by the yellow dye additives or the additives of anti-UV (that present in the film) penetrates and progresses in the thickness of film (Jin et al., 2006). Such UV penetration causes a progressive degradation not only on the surface but also through the thickness of the film.

Such observed ageing and deterioration in all mechanical properties (yield strength, fracture stress and strain, elastic modulus, resilience and toughness) of LDPE greenhouse cover is attributed to the climatic conditions such as solar irradiation, temperature, humidity, rain, and wind loads, and environmental pollution. UV radiation (290–400 nm) is known as the high energy part of the solar spectrum that can be absorbed by the polyethylene. This cause bond cleavage and polymerization, that leads to photodegradation (oxidation) and thus mechanical degradation. This has been reported by several investigators for mono-layer film (e.g., Khan and Hamid (1984) and Salem (2001)). Salem examined the effect of UV radiation on mechanical properties of mono-layer LDPE films containing black carbon titanium oxide. He has shown that the UV radiation has changed the elongation and shear stress of LDPE films. The degradation in all mechanical properties has been observed even after the first month of exposure to natural ageing as shown in Figs. 1–7. This is in contrast with the observation of Al-Madfa et al. (1998). They have shown that during the natural ageing, the mechanical properties of polyethylene are improved during the first months of exposure and that after they collapse significantly.

The findings of this work revealed that the mono-layer and tri-layer lost 50% of their mechanical properties after exposure to natural ageing of 5 months and 10 months, respectively. The study of Ram et al. (1980) showed that the unprotected mono-layer LDPE films failed (50% retained elongation) after the equivalent of < 2 months exposure. Qureshi et al. (1990) have reported that linear LDPE (LLDPE) mono-layer films had a 50% reduction in tensile strength at break after only 3 months of natural exposure in a hot region.

To the background of the authors, there is not a study available in the literature addressing the effect of ageing on the greenhouse tri-layers LDPE cover except the work of the authors Dehbi et al. (Youssef et al., 2008a, b; Dehbi et al., 2010). As discussed in the preceding sections the tri-layers film is more resistive to ageing than the mono-layer one and consequently high lifespan.

Since the natural ageing induced sever degradation on the external surface of film due to the sunlight radiation which is a major element of degradation. Therefore, such high resistance of degradation of the tri-layers film can be interpreted based on the composite structure it is made with. It has been confirmed from the manufacturer that the co-extruded three layers are adhered together by heating to a temperature level of 70 °C. Such structure/design allows the forming of three boarders between each two mating surfaces. The migration of the additives such as (UV stabilizer, antioxidant and Nickel stabilizer) LDPE film along ageing is a crucial parameter in the degradation of LDPE film used as greenhouse cover, for the mono-layer the migration of the additives is easier than for the tri-layers, both upper layer blocks the additives in the intermediate layer and makes it possible to slow down the migration of the additives towards outside face in the tri-layers film. In another words, the rate of diminishing of the protective additives in the tri-layers film is slow than in the mono-layer film. This is the expected reason behind the large durability of the tri-layers film.

From economical point of view, it has been confirmed from the company that the costs of mono-layer and tri-layers films are 90 and 100 DZ/m^2, respectively. That is 11% more per metre. The greenhouse consumes about 250 m^2 which means that there is a difference of only 2500 DZ if the cover will be made of tri-layers film. Based on the results of this work from both technical and economical perspectives, the tri-layers film is highly recommended for using as a cover for the greenhouse. Such findings highly motivated the authors and open the door to study the performance/durability of the five-layers and compare with the tri-layers films.

4. Conclusions

The influence of different exposure times (up to 5040 h/7 months) of natural ageing on the mechanical behaviour of mono-layer and tri-layers LDPE films used as greenhouse cover was investigated. The tensile tests have been conducted to explore the mechanical properties (yield strength, elasticity modulus, fracture stress, strain to break, resilience, toughness) of version and aged films. The mechanical properties of both films were deteriorated due to the exposure to the environmental conditions (e.g., solar radiation and temperature). The sunlight radiation is the major element of degradation. The durability of both LDPE mono-layer and tri-layers films is discussed in detail in this article. The results of this work revealed that the degradation of the mechanical properties is interrelated to the weathering and ageing process. As far as the resistance to weathering and to the effect of UV radiation, high temperature, etc., is concerned, tri-layers cover is considered to be inherently more stable and can withstand ten months exposure without exceeding the 50% deterioration in the mechanical properties. On the other side the mono-layer film deteriorates rapidly within only five months on continuous exposure to ageing. There are marked decreases in mechanical properties leading ultimately to disintegration. The design of tri-layers film makes it possible to slow down the migration of the additives towards outside face and consequently make it more durable and has more lifespan. Based on the mechanical characteristic, lifespan criterion and cost analysis, the tri-layers cover is highly recommended than mono-layer film as a greenhouse cover.

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