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CLIMATIC AGEING OF LOW DENSITY POLYETHYLENE IN AGRICULTURAL GREENHOUSES

ABSTRACT

Low-Density Polyethylene (LDPE) is a commonly employed plastic in the production of greenhouse covers due to its advantageous attributes of flexibility, durability, and transparency. Notably, LDPE's ability to resist UV radiation plays a crucial role in protecting plants from harmful solar rays. However, the extended lifespan initially anticipated for this material is frequently undermined by the intricate process of LDPE aging. This study examines how the mechanical properties and creep behavior of monolayer and tri-layer PE materials change over nine months of natural aging in the Tiaret region of northwest of Algeria. Tri-layer polyethylene (PE) film outperforms mono-layer PE in strength (2.37x), elasticity (35%), and creep resistance due to its enhanced barrier against environmental factors. It maintains 50% elongation even after 9 months, while mono-layer PE degrades 50% in six months with a tensile strength of 14 MPa.

Keywords: *LDPE; ageing; lifetime; mechanical properties; creep*

INTRODUCTION

Polymer materials have become a crucial aspect of our society, with a wide range of applications in different fields, depending on their particular types and properties. Polyethylene (PE) and polypropylene (PP) are two of the most commonly used polymers for films due to their low cost, flexibility, durability, and easy processing [1-3]. Ethylene copolymers with vinyl acetate or vinyl alcohol, ethylene copolymers with methacrylic acid, and polyvinyl alcohol are also commonly used as they offer improved properties such as increased clarity, adhesion, and moisture resistance. Fluoropolymers, such as polytetrafluoroethylene (PTFE) and fluorinated ethylene propylene (FEP), are also used for films in applications where high temperature resistance and chemical resistance are required.

The annual consumption of plastic material in the agricultural sector is approximately 5 million tons, which accounts for about 2% of the total plastic production worldwide. This includes a wide range of plastic products such as greenhouse films, mulch films, irrigation pipes, packaging materials, and other plastic products used for various agricultural purposes.

Knowledge of the characteristics of a polymer is essential when choosing its use in a given field. The variable climatic conditions have an impact on their physico-chemical properties which will cause their harmful degradation (chemical degradation) and (physical degradation) [3,4]. Thereby, natural (climatic) ageing is the result of exposure of the material to various climatic agents, such as: Solar radiation, day/ night temperature gradients, wind strength variations, water (moisture), snow, hail and sand [5-10]. These factors determine the polymers' useful life and their economic and environmental impact [11]. Sunlight is the primary detrimental factor in the ageing of greenhouse [12].

Ageing, which represents a form of degradation of polymers, corresponds to a slow and irreversible evolution of one or more properties of the material from a reference point, generally taken at the end of the manufacturing cycle. Nevertheless, the degradation mechanism of PE is yet not well understood [13]. Polyethylene (PE) environmental degradation is a complex process, as many degradation mechanisms act together toward the total destruction of the material.

The greenhouse films used in plasticulture are intended to create adequate climatic conditions to obtain better control of the cultivation conditions [14]. Indeed, their fundamental function is to protect the crop from bad weather, in particular hail and cold. They are used to optimize growing conditions (light, temperature, and humidity) and create an ideal environment for plant growth [15]. Further, the function of the greenhouse cover is dependent to a far extent on its optical properties in addition to its mechanical ones [16,17]. These films may be single or multi-layer. In the second case, the film consists of different layers of materials, thanks to which it combines the features of components.

Using single layers of polyethylene (PE) in agricultural greenhouse construction is simpler and cheaper than using triple-layer structures. However, potential losses due to poor barrier properties and durability of single-layer PE may lead to increased waste and financial setbacks. Triple-layer films offer better protection against moisture, oxygen, and light, leading to improved product preservation, reduced deterioration, and waste reduction. This benefits the environment by conserving resources. As technology advances and demand for eco-friendly plastics grows, the cost difference between single and triple layers is likely to decrease. Choosing triple-layer plastics becomes more attractive economically and environmentally.

A significant progress and several future directions for polymer materials in agricultural films can be anticipated namely: the Integration of biodegradable polymer elements, the design of intelligent film, the inclusion of nanoparticles within polymer matrices, the development of films conducive to simplified recycling processes, the creation modified atmosphere films, the design of specific films to offer increased protection against UV radiation and using biopolymers derived from plant or microbial sources [18-20].

Many researchers were interested in long term behavior of polyethylene films. Dehbi and Mourad [1,16] examined the effects of natural aging on a mono-layer and tri-layer LDPE film over a period of 7 months. The results revealed that the mechanical, optical properties, durability and lifespan of the tri-layers films are found to be quietly better than that of the mono-layer films. Benmiloud et al. [6] have checked by infrared spectroscopy the microstructural and morphological changes, and have followed by tensile and creep-recovery tests the changes of the mechanical properties on the films naturally aged for 6 months. They conclude that the improvement of the creep resistance affects the different deformations and the short chain segments coming from chain scissions increase the crystallinity ratio, and lower each of the strain rate. Dehbi et al. [9] have studied the degradable effect of natural and artificial ageing on tri-layer of low density polyethylene (LDPE) films. The study has been carried out over a period of eight months of natural and artificial ageing. The films have been

artificially aged at 40°C, 40°C + UVA, 50°C and at 50°C + UVA. The findings show that the environmental factors and especially the simultaneous effect of temperature and UVA radiation have degradable effects on the durability and all mechanical properties of the polyethylene film.

Yousef et al. have studied the effect of natural aging on the tri-layer LDPE films [17,21] during seven months. They have shown that the trilayer film duration of use was estimated to be 10 months and the structural change occurs on the external surface of the films, while the interior surfaces are protected by the dye. The effect of sand wind at 40°C for 4 h decreases the optical transmission of a single layer of LDPE by 50%.

Djakhdane et al. [22] have studied the degradable effect of artificial ageing on tri-layer LDPE films over a period of one to eight hours. Optical, thermal, surface analysis and mechanical properties have been analyzed on samples having undergone different thermal treatments associated with sand and wind simulation at 40°C in order to test their performance when used in a Saharan environment. The results revealed that the ageing conditions affected adversely the mechanical behavior of the film and the severest condition was for ageing at 40°C with sand-wind for 8 hours.

Temperature variations are very significant in the Tiaret region (Algeria). Temperatures are variable according to the seasons but also between day and night. Indeed, the winters are very cold, the temperature sometimes reaches −5°C. While in summer, the thermometer sometimes shows 45°C in the shade. The annual average temperature in Tiaret is below 18°C.

Unlike temperature, which usually varies considerably between day and night, humidity levels in Tiaret vary by days but at a level of humidity qualified as heavy or stuffy, cannot exceed 1%; it is therefore an insignificant value compared to those recorded in coastal regions throughout the year [23].

The main property used by industry for characterizing the degradation of PE is the elongation at break. According to relevant international standards, a lowering below the limit of 50% of elongation at break compared with the value at new is the indicator that the material has finished its working life and has to be removed before its mechanical characteristics [2,24-28].

This article aims to investigate and optimize the degradation induced by weather ageing of a commercial grade low density polyethylene film (LDPE), mono-layer, and tri-layer used in agricultural greenhouses. Two authentic greenhouses of the same dimensions, made from mono and triple-layer polyethylene, have been installed at a vacant site located in an arid zone in Tiaret in the northwest of Algeria. Three samples of mono-layer and tri-layer plastic films of well-defined dimensions are collected after each month over a duration time of nine months (6480 h) and analyzed by tensile and creep tests. The values given in this article represent their average. The load was applied to the specimen in a direction parallel to the average molecular orientation obtained during the film processing.

MATERIALS AND TESTS CONDUCTED

Materials characterization

The monolayer (PEbd) and the tri-layer (PEbd) AFS3 were obtained from Agro-Film company, Sétif, Algeria. The raw LDPE mono-layer (before extrusion) used in this study has the characteristics indicated in Table 1.

Table 1. *Monolayer characteristics*

Parameters	Density (g/cm^3)	Mono-layer thickness (μm)	Molecular weight (g/mol)	MFI (g/10 mn)
Values	0.923	180	90000 - 120000	0.33

MFI: The melt flow index

The thickness of the three co-extruded layers film an extrusion temperature of 70°C is 180 μm with the proportions of 1/4, 1/2, and 1/4 in the layers. That is the same thickness as the mono-layer film (Figure 1).

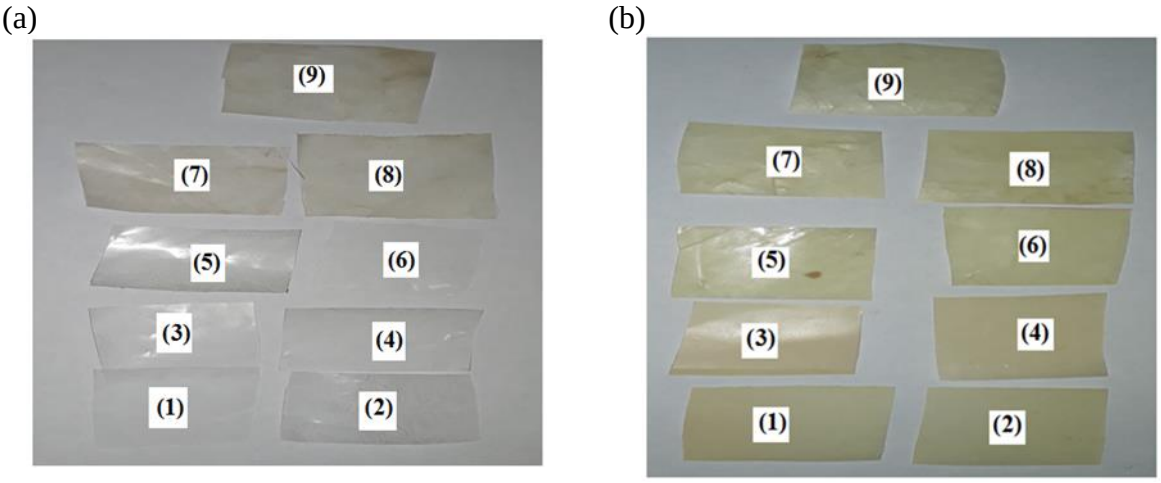

Fig. 1. Polyethylene films: (a) mono-layers (collected after each month); (b) tri-layers (color of the film is milky yellow)

In the present work, polyethylene films mono-layers and tri-layers used as greenhouse cover in the region of Tiaret / Algeria have been weathered in severe climatic conditions.
These conditions (temperature and moisture average) have been tacked at 00 pm, and 14 pm during May to January (9 months = 6480 h) as shown in Figure 2.
It should be noted that between June and October, the thermometer displays these maximum temperatures. They are between 26 and 35°C during the day and between 18 and 25°C at night. The maximum moisture is 45-70% during the day, for the months of May, November, December and January. It is 70 to 90% at night, for the same months.

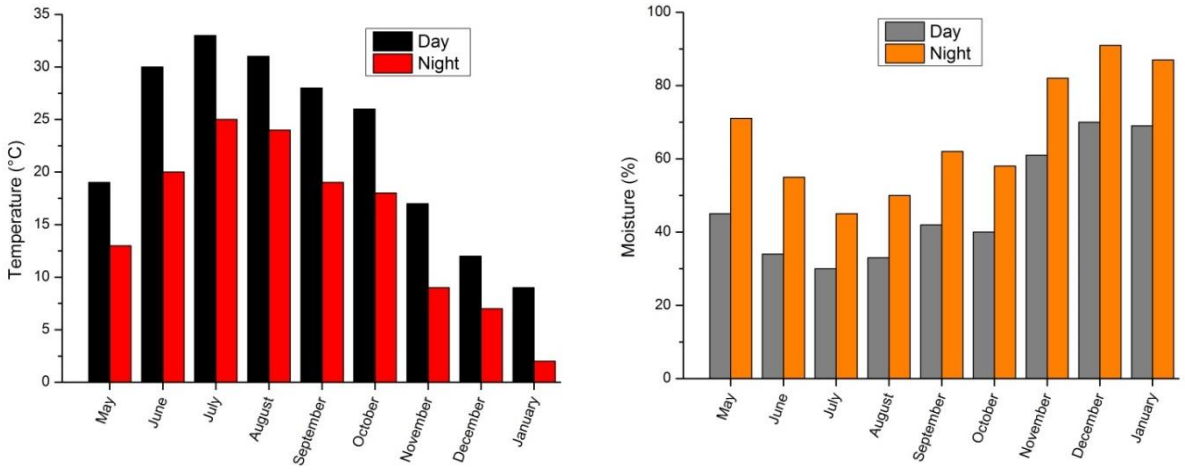

Fig. 2. Temperature (left) and humidity (right) sampled from May to January in the Tiaret region / Algeria

Tensile test specimens

The geometry and dimensions of the tensile specimens (Figure 3) were produced in accordance with standard NF EN ISO 527-3. All the tests were carried out at room temperature with a tensile force of 5 kN and a tensile speed set at 50 mm/min.

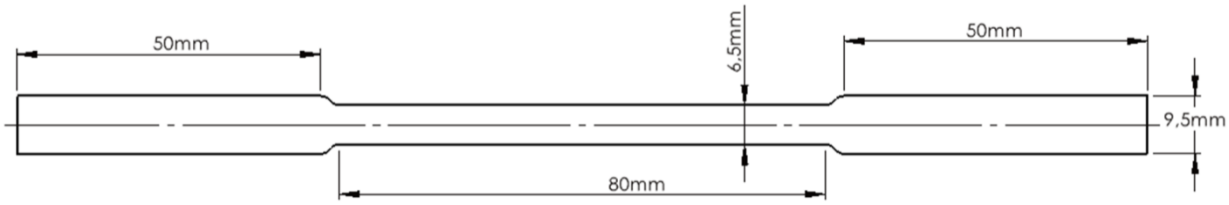

Fig. 3. Geometry of tensile specimens

Creep test specimens

Creep is the deformation of a material with time when the material is subjected to a constant load for an extended period of time i.e. time-dependent deformation. The change of strain can lead to considerable reduction in life expectancy of the component. Strain (or deformation) increases with load, temperature, relative humidity and time.

The creep test has been performed with rectangular specimens cut out of the virgin and aged film during 1, 2, 3, 4, 5, 6, 7, 8 et 9 months. Their geometry and dimensions are shown in Figure 4. The creep tests were conducted by applying a constant load delivering an initial stress of 7.6 MPa (i.e., 1500 g) for duration of 24 h by specimen. The recovery starts immediately after the stress removal and for a period of 24 h as well.

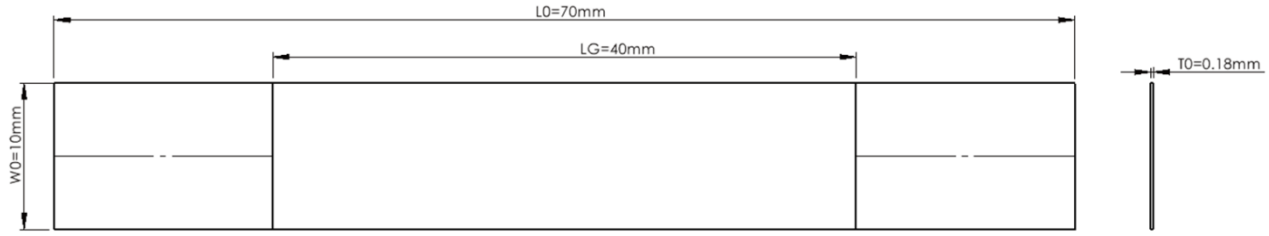

Fig. 4. Creep test specimen geometry

RESULTS AND DISCUSSION

Strength, ductility, and the deformation of the material's time-dependent testing were conducted during 9 months of the mono-layer and tri-layer LDPE films. The experimental results are displayed below:

Elongation at break

Comparing the elongation at break of the mono-layer LDPE film and a tri-layer LDPE film involves evaluating their respective capabilities to elongate before failure.

The elongation at break decreases continuously with time. For the mono-layer LDPE film ranges from 85 % for the first month to 20% for the ninth month. The elongation at break of the tri-layer LDPE film ranges from 190% to 120% from the first month to the ninth.

The ratio of the elongation at break of the tri-layer film to the mono-layer film is about 3.5 during the 9 months of exposure. This means that when the number of layers is multiplied by 3, the average elongation at break of the three-layer film increases by a factor of 3.50 (Figure 5).

Tri-layer films have higher elongation than single-layer films because the extra layers prevent the film from becoming brittle or prone to cracking during elongation due to better barrier performance and more even stress distribution. The orientation of the polymer chains [1,10,16], their molecular weight [2,18], the degree of crystallinity [2,6,10], the temperature, and the humidity [3,16,29,30] can influence the elongation property of LDPE.

All authors agree in affirming that the deformation at rupture (elongation at break) is the mechanical parameter that best represents the state of material degradation, which has led to the introduction of the concept of lifetime as the exposure time required for it to reach 50% of its initial value [22,24-28]. In regards to the mono-layer PE, it reaches its half-life after six months of exposure, corresponding to a tensile strength value of approximately 14 MPa. Unlike the tri-layer PE, the 9 month exposure time to the natural conditions were insufficient to cause a loss of 50% in the elongation at break.

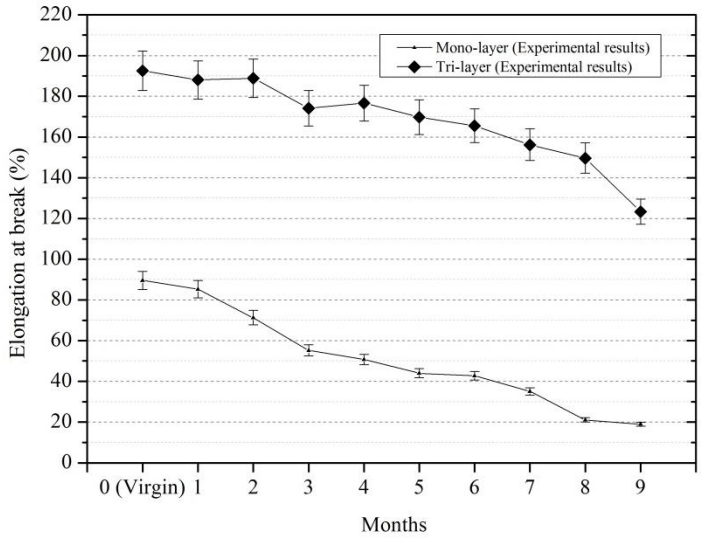

Fig. 5. Comparison between the elongation at break of the monolayer and the tri-layer

Breaking stress

The breaking stress values of the tri-layer material significantly outperform those of the monolayer after 9 months of aging. This difference in stress lies in the cooperative effect of interlayer bonds in multilayer structures. However, these values gradually decline as the aging time increases. The average deviation of the values of breaking stress after aging for 9 months is 3 MPa for tri-layer and 1.8 MPa for monolayer (Figure 6).

For the three-layer, a greater drop in the value of the breaking strength is to be taken between month 3 and 4 and between month 8 and 9 corresponding to very high values of temperature and humidity respectively.

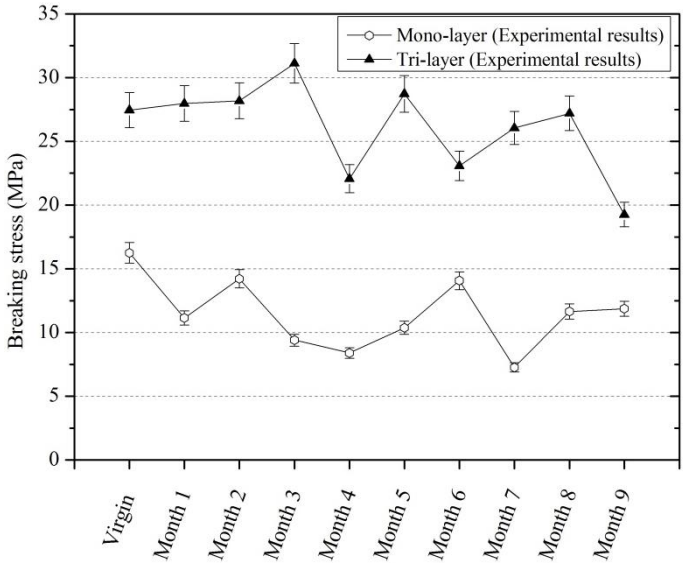

Fig. 6. Breaking stress of monolayer and tri-layer

Trilayer films exhibit a significantly greater breaking stress compared to monolayer films, with a strength that is 2.37 times higher.

The enhanced strength and mechanical properties of trilayer films can be attributed to the degree of crystallinity of LDPE [10,31], and to the combination of materials and their interfaces. By stacking three layers, trilayer films can benefit from the synergistic effects and efficiently distribute stress throughout the layers, making them more resistant to failure.

Elastic stress

The elastic stress for mono-layers and tri-layers is illustrated in Figure 7.

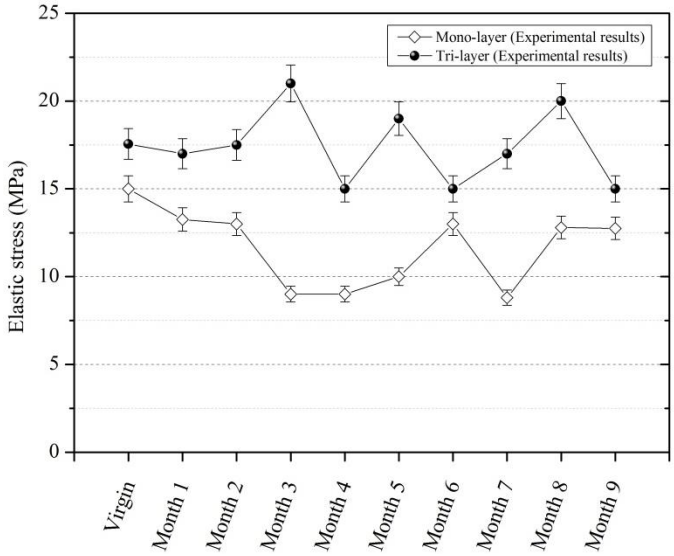

Fig. 7. Elastic stress of monolayer and trilayer as a function of the number of months

The elastic limit undergoes significant variations over time. The average deviation varies between 1.7 and 1.9 MPa. The elastic stress values of the tri-layer material are higher than those of the monolayer due to the thickness which can provide material reinforcement and bonding between layers which influences significantly these values. The tri-layer material exhibits elastic stress values that are 1.54 times higher than those of the monolayer material. This means that the tri-layer material is experiencing 35% more elastic stress compared to the monolayer material. The degradation in the elastic stress of the mono-layer film of LDPE is due to the changes occurring with ageing in the molecular structure of the material [19,22,30].

Comparing the outcomes of this study to those presented in reference [1], the authors in reference [1] examined the effects of aging on a mono-layer and tri-layer LDPE film over a period of 7 months. Both our results and those obtained demonstrate that trilayer films exhibit mechanical properties that surpass those of monolayer films. However, the values measured for these properties in this study are considerably lower than those reported in reference [1]. This disparity can be attributed to the quality of the LDPE films used in their research, which contained various additives.

Strain rate

Figure 8 illustrates the relationship between the strain rate of the tri-layer and the corresponding time.

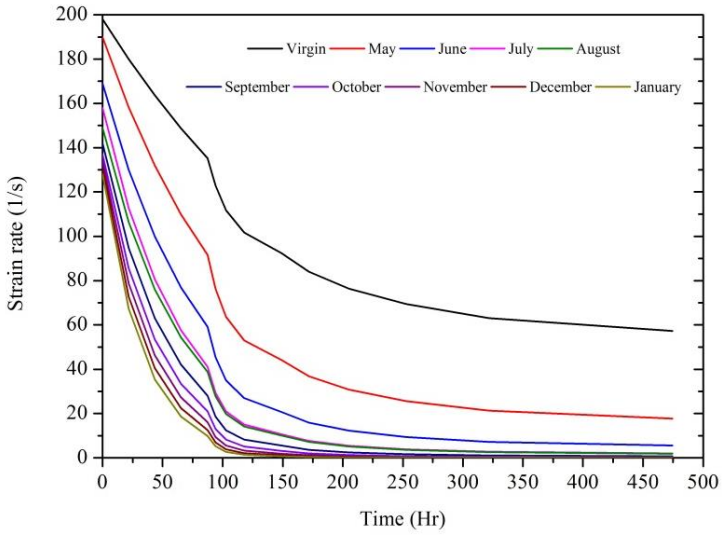

Fig. 8. Strain rate of the tri-layer polyethylene films

The strain rate decreases for the tri-layer as the number of months increases, which leads to a decrease over time in the ability of the material to deform under stress. This behavior is attributed to ageing of the molecular structure [2] and the material's degradation with time through an accumulation of microstructural defects and changes in the material's composition.

Correlation between average temperature and minimum strain rate (MSR)

Figure 9 expresses the relationship between the average temperature between the day and the night of the nine months of aging and the minimum strain rate (MSR).

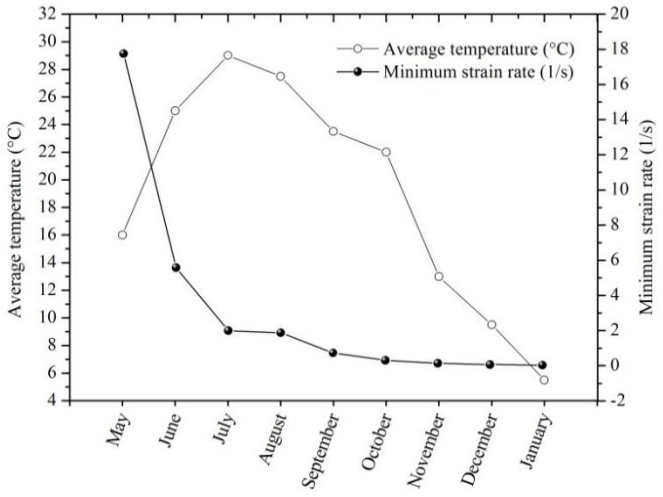

Fig. 9. Correlation between average temperature and minimum strain rate (MSR)

It is noted that for polyethylene films, as the temperature increases, the strain rate also tends to increase. This is because higher temperatures facilitate molecular mobility, allowing the polymer chains to move more freely and thus leading to easier deformation and higher strain rates.

From the month of July, the minimum strain rate decreases with a decrease in temperature. This drop in temperature causes the thermal energy to decrease, reducing the mobility of the atoms or molecules within the material, and do not allow the chains to re-orient and provide additional resistance following the initial yield of the material [5,8,9]. As a result, the material becomes less prone to deformation, and the strain rate tends to decrease.

Correlation between Tr and MSR for the LDPE Trilayer

The correlation between the rupture time (Tr) and minimum strain rate (MSR) of the LDPE trilayer is shown in Figure 10, expressed as:

$$\mathrm{Tr} = 2494.7\mathrm{e}^{-0{,}103\mathrm{MSR}} \tag{1}$$

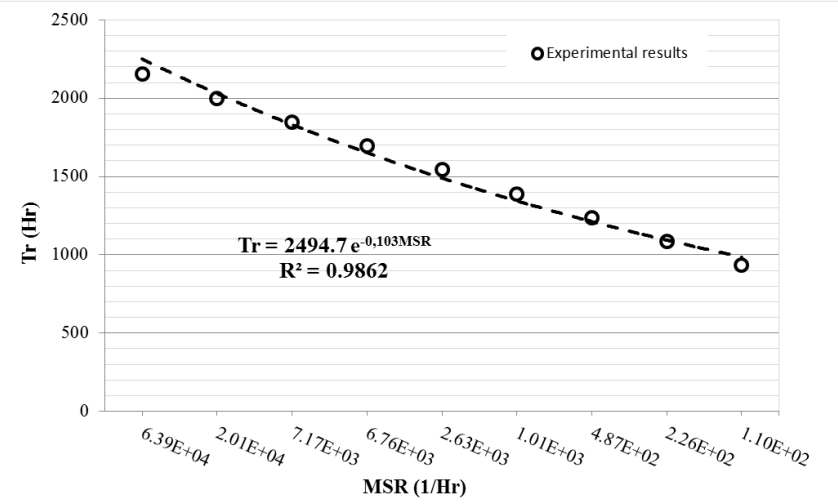

Fig. 10. Correlation between Tr and MSR for the LDPE Trilayer

Trilayer LDPE displays an increase in minimum strain rate, resulting in an increase in strength and resistance to deformation, and extended rupture time.

This increase in minimum strain rate is associated with an increase in strength, which means that the trilayer LDPE film can withstand higher forces or stresses without breaking or deforming permanently. It also contributes to the film's resistance to deformation, making it more durable and less prone to tearing or puncturing.

Moreover, the extension of the film's rupture time implies that it takes longer for the trilayer LDPE film to reach its breaking point when subjected to stress. This characteristic is beneficial in various applications, such as agricultural greenhouses, where the film needs to maintain its integrity and protect the agricultural products for an extended period.

CONCLUSIONS

This study aimed to investigate the impact of natural aging on the mechanical characteristics of mono-layer and tri-layer LDPE films. To achieve this, monotonic tensile and creep tests were conducted on both aged and unaged specimens. The aging process involved exposing the samples to natural thermal aging conditions to replicate the effects of prolonged environmental exposure or service life. Based on the experimental findings, the following conclusions can be made:

- The study has effectively achieved its goal, leading to both qualitative and quantitative evaluations of the tensile and creep mechanical characteristics of trilayer and monolayer films.
- The tear strength and the elastic limit (yield point) undergo slight variations over time, while the elongation at break and the tenacity decrease in an accentuated way for the samples aged for a longer time.
- The tri-layer film shows a 75.55% increase in mean elongation at break compared to the mono-layer material.
- The mono-layer LDPE reaches its half-life after six months of exposure, corresponding to a tensile strength value of approximately 14 MPa. Unlike the tri-layer LDPE, the 9 month exposure time to the natural conditions were insufficient to cause a loss of 50% in the elongation at break.
- Trilayer films demonstrate a remarkable increase in breaking stress, surpassing monolayer films by 57.88%. Additionally, they exhibit a substantial 35% increase in elastic stress compared to monolayer films.
- Primary and secondary creep are observed in all creep curves, whereas tertiary creep is absent because the creep test is carried out at low temperatures and stresses.
- Utilizing tri-layer LDPEs presents an enhanced and environmentally-conscious approach for lasting effectiveness, achieved through the integration of biodegradable polymer elements, intelligent film design, the inclusion of nanoparticles within polymer matrices, and the development of films conducive to simplified recycling processes. ...and so on.

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Data availability statement
The data that support the findings of this study are available on request from the corresponding author.
Conflicts of Interest
The authors declare that there is no conflict of interest.

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