JMEPEG (2006) 15:111-116
DOI: 10.1361/105994906X83475

1059-9495/$19.00

Viscoelastic Behavior of HDPE Polymer using Tensile and Compressive Loading

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(Submitted July 8, 2005; in revised form September 7, 2005)

Large containers for liquids, exposed to different static loadings, are mainly made of high-density polyethylene (HDPE). The viscoelastic response of HDPE under long-term tensile and compressive creep is investigated. Monotonic experiments under tension are performed over a wide range of strain rates. In these experiments, the transition in the damage mechanisms (development and propagation of contraction in the HDPE specimen) is analyzed. The monotonic tensile behavior of the HDPE is found to be nonlinear and depends on the strain rates. It is observed that both elastic modulus and plastic flow stress present an increase with displacement speed due to the viscoelastic behavior of HDPE. A similar observation can be made for monotonic compressive tests by developing a new experimental device that ensures accurate measurement of the strain. Such a device makes use of an extensometer of compressive displacement of the specimen. In addition, the long-term behavior of HDPE is evidenced through creep and relaxation tests at an imposed range respectively of lower stresses and strains. It is shown that the normalized curves, associated with these tests, can be represented by a single curve characterizing the compressive creep compliance or relaxation stresses versus time. The linearity of the viscoelastic behavior is confirmed within the linear domain of the monotonic compressive and tensile tests.

Keywords compression, creep, high-density polyethylene (HDPE), relaxation, tensile

1. Introduction

Plastic containers and bottles have found widespread application for storage of consumer products and such industrial chemicals as alcohols and acids. High-density polyethylene (HDPE) is frequently the choice for such containers, especially for those liquids that are stored at atmospheric pressure (Ref 1-3). HDPE combines low cost, excellent properties, and good toughness (Ref 4, 5). In addition to its appealing mechanical properties, HDPE is a very good candidate for recycling (Ref 6-8). Stacked HDPE containers of liquids are subject to static loading and strain rate deformation. Generally, polymers are known to show different mechanical responses under tension and compression in static loading conditions (Ref 9). Knowledge of the long-term response of these materials is essential for developing a good understanding of their mechanical behavior (Ref 10-12). Data collection from experiments for simple and complex loading paths is increasingly needed with the growing use of polymers in various fields (Ref 13). Polymeric responses under impact have been studied. Chen et al. (Ref 14) established quasi-static experiments under both tension and compression testing on poly(methyl methacrylate) (PMMA) to determine stress-strain responses at various strain rates. Moreover, a study on stress relaxation was performed on two grades of polycarbonate (PC) (Ref 15).

Thanks to the simplicity of tensile tests, a large number of studies on tensile creep experiments for many polymers, in particular HDPE, are still in progress (Ref 16-20). Because HDPE materials exhibit time-dependent viscoelastic mechanical behavior, a thorough knowledge of such behavior is essential for optimum design (Ref 20). It is commonly known that HDPE is an important material whose tensile creep is a good measure of its viscoelastic nature (Ref 21). However, compression tests have rarely been addressed in the literature due to the difficulty of measuring directly the deformation of the specimen. Because HDPE, used in bottles and containers for liquid manufacturing, is known to be under permanent loading, it is necessary to conduct compression tests.

The objective of this study is to describe the viscoelastic behavior of HDPE under tensile and compressive loading. For this, compression tests on HDPE are carried out using a universal tensile testing machine equipped with a newly developed device for direct measurement of specimen deformation. These tests are conducted in two steps. First, monotonic tensile and compressive tests are performed at different strain rates to characterize the mechanical properties of HDPE and, thus, to determine the most suitable strain rate for future tests. In these tests, transition in the damage mechanisms is analyzed. Second, the long-term behavior of HDPE is evidenced through tensile and compressive creep tests at constant, lower loads.

2. Materials and Methods

2.1 Sample Preparation

The polymer of interest is a high-density polyethylene (B5429). It is a linear polymer with the chemical composition $(CH_2)_n$. It is made of Deutsche Sammlung von Mikroorganismen (DSM) as a product of ethylene polymerization with a density of 0.93 g/cm^3 and a Shore hardness of 96 (Shore A). Differential scanning calorimetry (DSC) was used to determine the glass transition T_g of HDPE: –75 ± 3 °C. Compression tests

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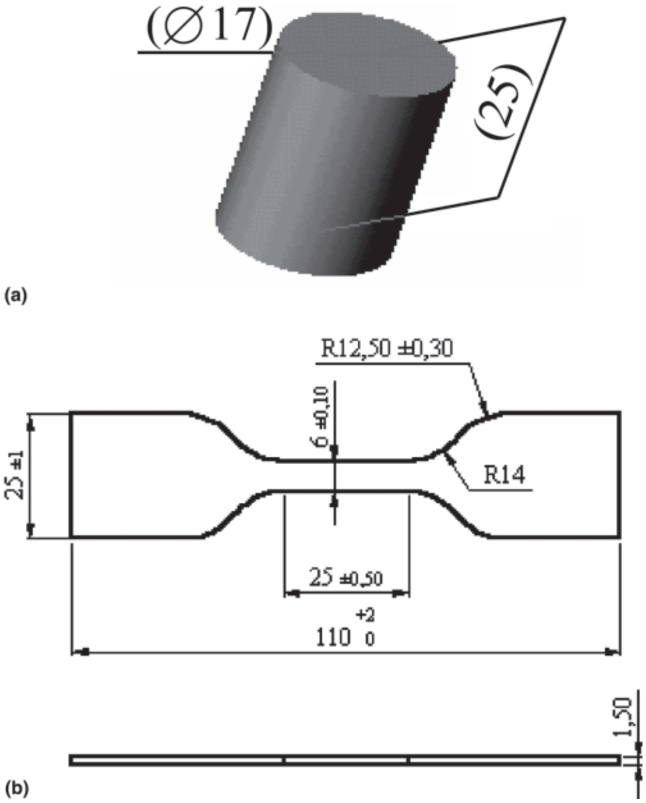


Fig. 1 (a) HDPE specimen for compressive test; (b) HDPE specimen for tensile test

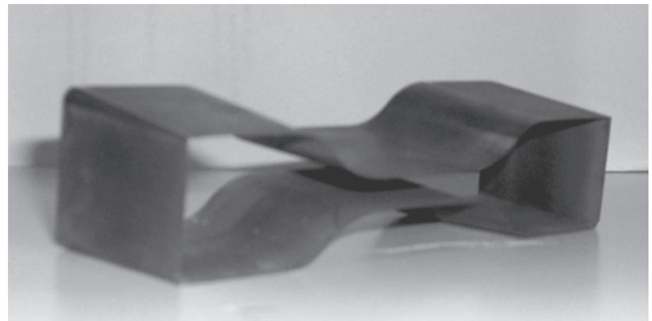


Fig. 2 Developing cutting tools

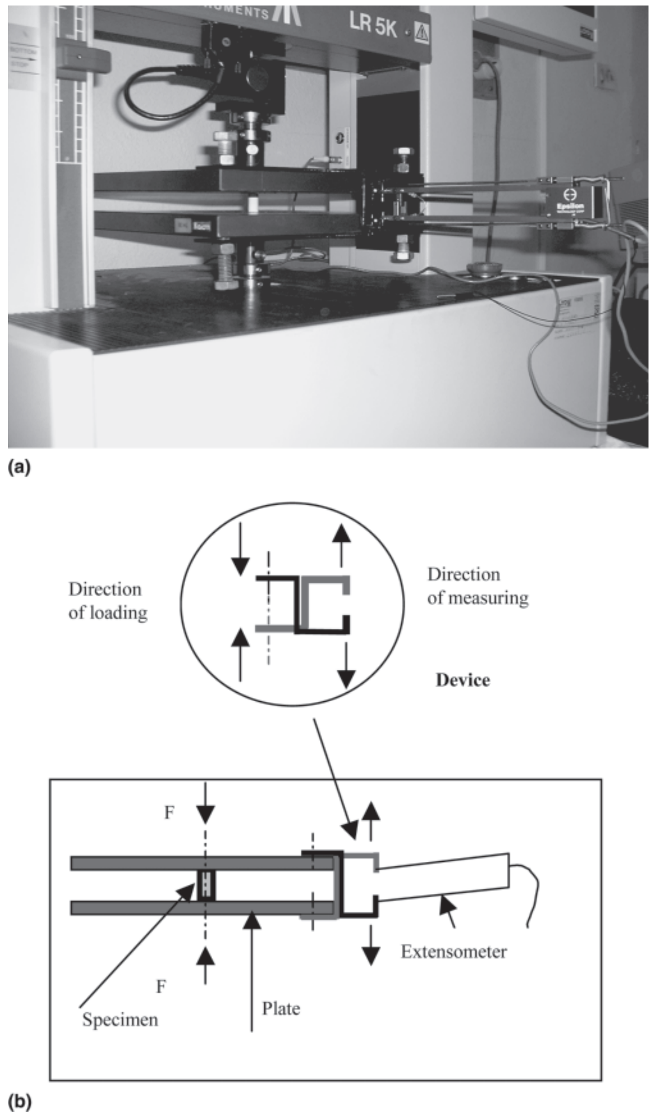


Fig. 3 (a) Compressive test; (b) schematic representation of the compression test procedure

were performed on cylindrical samples shown in Fig. 1(a). Specimens were prepared directly from the extrusion head, the diameter (ϕ) and height (h) of which are 17 and 25 mm, respectively. Specimens (Fig. 1b) for tensile tests were obtained by cutting samples from the container using a cutting tool (Fig. 2), which is fabricated with the aid of CNC wire cut electrical discharge machining (EDM) to ensure that the samples' dimensions are of high precision. For each experiment, a minimum of five samples were tested to estimate the mean value and obtain the standard deviation.

2.2 Tensile and Compression Testing

Tensile tests were conducted on a standard traction-compression machine piloted in displacement mode using an extensometer to measure the deformation of the specimen. The output of the measured deformation and load were continuously stored using a PC for data acquisition. With the aid of the same machine, compression tests were performed. To have a direct measurement of the compression displacement on the specimen, a new experimental device was developed. This device was attached to the parallel plates of the universal tensile testing machine to use an extensometer. This apparatus eliminates errors introduced by the load frame deformation and the drive system, and therefore, a precise measurement for direct deformation on the specimen during compression is obtained (Fig. 3a, b). To minimize the friction coefficient between the contacting surfaces (the contact between the specimen and plates), surfaces were lubricated with oil. The specimen was positioned at the center of the plates. Tests were carried out at the displacement rates of the machine frame, 6, 60, and 600 mm/min, corresponding to the strain rates of the tensile and compression tests $\dot{\varepsilon} = 4 \times 10^{-3}\ s^{-1}$, $\dot{\varepsilon} = 4 \times 10^{-2}\ s^{-1}$, and $\dot{\varepsilon} = 4 \times 10^{-1}\ s^{-1}$, respectively. In these tests, the temperature was measured at 25 ± 1 °C and the relative humidity (RH) was found to be 40 ± 10%. With regard to the tensile and compressive creep experiments, constant low stresses $\sigma_0 = 3$ MPa, $\sigma_0 = 5$ MPa, and $\sigma_0 = 10$ MPa, in the linear range of stress-

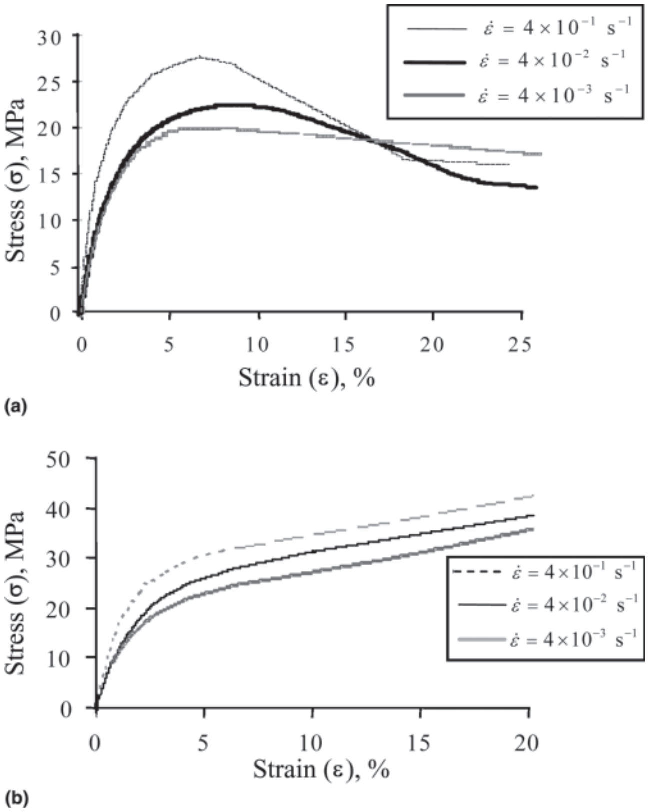


Fig. 4 (a) Monotonic tensile tests of HDPE: influence of strain rates; (b) Monotonic compressive tests of HDPE: influence of strain rates

strain curve, were applied to specimens at a constant strain rate ($\dot{\varepsilon} = 4 \times 10^{-2}$ s^{-1}). For the compression stress relaxation, however, tests were conducted on each sample with low applied strains of $\varepsilon_0 = 0.25\%$, $\varepsilon_0 = 0.41\%$, and $\varepsilon_0 = 0.83\%$, and for the tensile stress relaxation, tests were conducted on each sample with low applied strains of $\varepsilon_0 = 0.28\%$, $\varepsilon_0 = 0.47\%$, and $\varepsilon_0 = 0.94\%$, corresponding respectively to the aforementioned low stresses.

3. Results and Discussion

3.1 Monotonic Characterization of HDPE using Tensile and Compressive Tests

3.1.1 Influence of Strain Rates on Monotonic Tests. To examine the monotonic tensile and compressive response of the HDPE, it is necessary to assess the contribution of viscoelasticity in its behavior through the influence of the strain rate on its mechanical properties. In this study, testing speed needs to be determined by the specification of HDPE materials, and therefore, tests were carried out at a different range of strain rates. Three strain rates were chosen: $\dot{\varepsilon} = 4 \times 10^{-3}$ s^{-1}, $\dot{\varepsilon} = 4 \times 10^{-2}$ s^{-1}, and $\dot{\varepsilon} = 4 \times 10^{-1}$ s^{-1}. Typical stress-strain curves at different strain rates under tensile and compressive tests are displayed in Fig. 4(a) and (b), respectively. It is observed that the mechanical behavior of the HDPE at a wide range of strain rates is nonlinear and depends on the strain rates. Thus, it would be of great interest to analyze such an observation.

The elastic tangent modulus E (slope at the origin of the stress-strain curve) and the plastic flow stress σ_p (the stress at the beginning of the non linear part of the stress-strain curve)

Table 1 Mechanical properties of HDPE under compression tests at different strain rates

Strain rates, s^{-1}	Plastic flow stress (MPa)	Elastic modulus (MPa)
4×10^{-3}	14.4 (1.5)	1152 (54)
4×10^{-2}	15.4 (1.8)	1208 (60)
4×10^{-1}	18.5 (3.6)	1743 (60)

Table 2 Mechanical properties of HDPE under tensile tests at different strain rates

Strain rates, s^{-1}	Plastic flow stress (MPa)	Elastic modulus (MPa)
4×10^{-3}	13.3 (1.2)	978 (78)
4×10^{-2}	15.6 (1.5)	1060 (70)
4×10^{-1}	26 (3.6)	1523 (58)

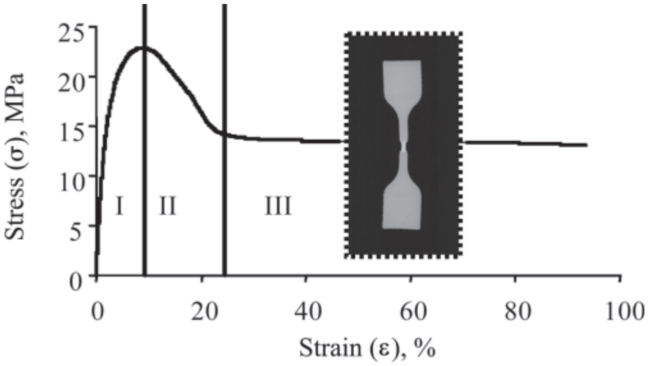
are presented in Tables 1 and 2. For tensile and compressive tests, at the low strain domain, the shape of the monotonic curves is similar and no significant difference behavior was observed for both $\dot{\varepsilon} = 4 \times 10^{-3}$ s^{-1} and $\dot{\varepsilon} = 4 \times 10^{-2}$ s^{-1}. In this domain, no distinction can be made between the mechanical properties at different strain rates up to $\dot{\varepsilon} = 4 \times 10^{-2}$ s^{-1}. Nevertheless, the original curve slope and stress yield behavior changed drastically at $\dot{\varepsilon} = 4 \times 10^{-1}$ s^{-1}. At high deformations associated with the monotonic tests, stresses increase in response to increasing the strain rates. Because the transition of mechanical properties occurs roughly at $\dot{\varepsilon} = 4 \times 10^{-2}$ s^{-1}, a similar strain rate is selected for the tensile and compression tests.

In conclusion, this study proves that the strain rates, in monotonic tensile and compressive tests, influences the HDPE behavior.

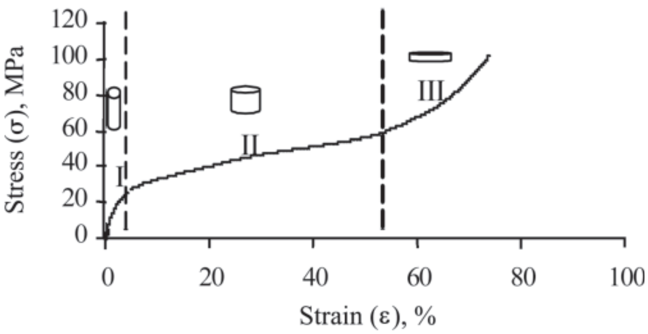
3.1.2 Transition in the Damage Mechanisms. Monotonic behavior of HDPE was investigated under tensile and compressive tests at $\dot{\varepsilon} = 4 \times 10^{-2}$ s^{-1}(Fig. 5a, b). Different damage mechanisms for both monotonic tests were observed. In these experiments, the transition damage (development and propagation of contraction in the HDPE specimen) was analyzed. From the experimental tensile curve of HDPE (Fig. 5a), three evolution stages are distinguished:

- Stage I is limited by maximum strength σ_{max} ($\sigma_{max} = 23$ MPa). This part is dominated by elastic behavior, and no change in the shape of the specimen was observed. The compressive elastic modulus (E), which corresponds to the slope at original tangent of curve, can be easily determined. It is approximated by 1060 (70 MPa).
- Stage II corresponds to a notable decrease of stress accompanied with a slight increase in the nonlinear deformation. At the strain value of 9.4%, the stress attains its maximal value denoted by the maximum strength (σ_{max}) and then decreases monotonically until the plastic flow stress (σ_p) corresponding to the strain value of 20%.
- Stage III is the plastic strain corresponding to stabilizing strain growth. At this stage, a development and propagation of contraction, over the entire calibrated zone of the HDPE specimen, was observed.

It is necessary to determine the loading and the strain to achieve creep and relaxation stress in tensile tests. Thus, low



(a)



(b)

Fig. 5 (a) Monotonic behavior of HDPE under tensile conditions; (b) monotonic behavior of HDPE under compression conditions

levels of stresses, $\sigma_0 = 3$ MPa, $\sigma_0 = 5$ MPa, and $\sigma_0 = 10$ MPa, corresponding respectively to the constant strains $\varepsilon_0 = 0.28\%$, $\varepsilon_0 = 0.47\%$, and $\varepsilon_0 = 0.94\%$, were selected in the linear domain for a monotonic stress-strain curve.

Three stages are also distinguished for the experimental compressive curve of HDPE (Fig. 5b).

- Stage I is characterized by a viscoelastic regimen at low stress in which the specimen geometry does not change. This zone is dominated by an elastic behavior with a compressive elastic modulus (E) of 1208 (60) MPa.
- Stage II corresponds to intermediate stresses associated with strains ranging from 3 to 55%. At this stage, a notable decrease in the curve slope can be observed.
- Stage III is identified by a yield behavior at high stress values and a change of curve orientation manifested by an increase in the curve slope. Eventually, the load continues to rise where the contact surface increases with load. The specimen becomes a thin disk, and thus, the sample loses its initial geometry and mechanical properties. With the newly shaped sample, failure of the specimen during the compression test was not possible.

It is also necessary to determine the loading and the strain to achieve creep and relaxation stress in compression tests. Thus, low levels of stresses, $\sigma_0 = 3$ MPa, $\sigma_0 = 5$ MPa, and $\sigma_0 = 10$ MPa, corresponding respectively to the constant strains $\varepsilon_0 = 0.25\%$, $\varepsilon_0 = 0.41\%$, and $\varepsilon_0 = 0.83\%$, were selected in the linear domain for a monotonic stress-strain curve.

3.2 Long-Term Mechanical Tests

3.2.1 Compressive Creep Behavior. Creep tests representing long-term testing were recorded during tensile and

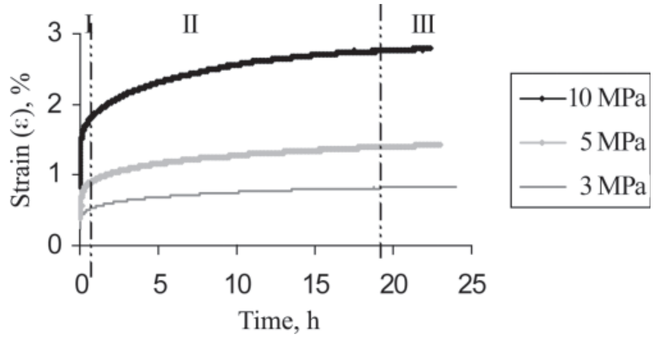


Fig. 6 Experimental compressive creep of HDPE versus time for different applied stress levels

compression tests at various imposed stress levels ($\sigma_0 = 3$ MPa, $\sigma_0 = 5$ MPa, and $\sigma_0 = 10$ MPa). The resulting material deformation of these tests is measured as a function of time (Fig. 6). The description of compressive creep curve at low stress levels exhibits three distinguishable phases:

- Stage I: a first creep deformation is the initial response, characterized by rapid strain rate where the material undergoes an important elastic deformation.
- Stage II: a longer time creep deformation in which HDPE undergoes a viscoelastic behavior.
- Stage III: a linear behavior of time-compressive creep is observed with very long deformation time.

Creep is characterized by a time function:

$$J(t) = \frac{\varepsilon(t)}{\sigma_0} \quad \text{(Eq 1)}$$

called "creep compliance" which corresponds to the strain per unit stress. $\varepsilon(t)$ is the time-dependent strain, and σ_0 is the applied stress. Using the time-dependent strain $\varepsilon(t)$ (Fig. 6), the creep compliance function $J(t)$ is determined at different imposed stresses: $\sigma_0 = 3$ MPa, $\sigma_0 = 5$ MPa, and $\sigma_0 = 10$ MPa. Figure 7(a) shows the variation of creep compliance versus time during creep experiments for different imposed compressive low stresses: $\sigma_0 = 3$ MPa, $\sigma_0 = 5$ MPa, and $\sigma_0 = 10$ MPa. Because the creep compliance curves are similar at low stresses, they can all be represented by one master curve. For the experimental tensile tests of HDPE, three distinguishable stages are also observed. In this case, the creep compliance curves can also be characterized by a master tensile curve (Fig. 7b). It can be concluded that, for both compression and tensile tests, the viscoelastic behavior of the HDPE at low stress levels is linear within the linear monotonic domain.

3.2.2 Compressive Stress Relaxation Tests. A similar observation can be made with regard to the stress relaxation under lower various constant strains ε_0. These strains are determined by the compressive stress σ_0 subject to:

$$\sigma_0 = \varepsilon_0 E \quad \text{(Eq 2)}$$

The long-term behavior of HDPE curves is evidenced through compressive stress relaxation tests versus time at lower strains ($\varepsilon_0 = 0.25\%$, $\varepsilon_0 = 0.41\%$, and $\varepsilon_0 = 0.83\%$; see Fig.

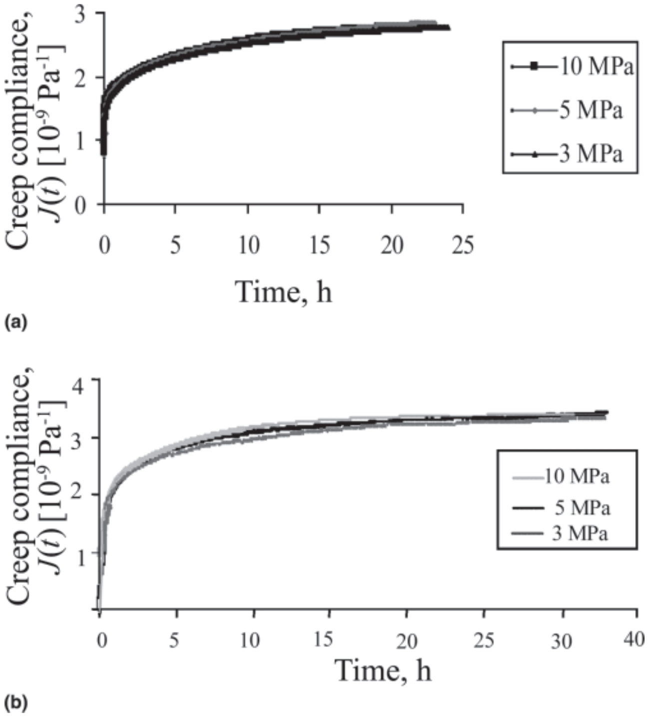


Fig. 7 (a) Experimental compressive creep compliance $J(t)$ of HDPE versus time for different applied stress levels $\sigma_0 = 3$ MPa, $\sigma_0 = 5$ MPa, and $\sigma_0 = 10$ MPa; (b) experimental tensile creep compliance $J(t)$ of HDPE versus time for different applied stress levels $\sigma_0 = 3$ MPa, $\sigma_0 = 5$ MPa, and $\sigma_0 = 10$ MPa

8). Notice that stress decays with time, and three stages are observed:

- Stage I: a quickly decreasing stress relaxation is characterized by short residence time. This decrease can be justified by lack of movement between chains.
- Stage II: a slowly deceasing stress to a constant value with possible macromolecular chains mobility (transition area) characterized by the relaxation time. This latter is defined as the time necessary for reorientation of chains under applied loads.
- Stage III: a constant stress characterizing a stabilized response.

The stress relaxation modulus $E(t)$ is defined as the stress per unit strain. It is given by:

$$E(t) = \frac{\sigma(t)}{\varepsilon_0} \qquad \text{(Eq 3)}$$

where $\sigma(t)$ is the time-dependent stress and ε_0 is the applied strain. The variation in the stress relaxation modulus versus time is shown in Fig. 9(a) at different lower values of applied strains ε_0. Similarly, the long-term behavior of HDPE curves are evidenced through tensile stress relaxation tests versus time at lower strain ($\varepsilon_0 = 0.28\%$, $\varepsilon_0 = 0.47\%$, and $\varepsilon_0 = 0.94\%$). The variation of the stress relaxation modulus $E(t)$ versus time is displayed in Fig. 9(b).

The superposition of the relaxation modulus $E(t)$ of HDPE, for different applied strains in compressive and tensile tests (Fig. 9a, b), confirms that the relaxation curve is unchanged in

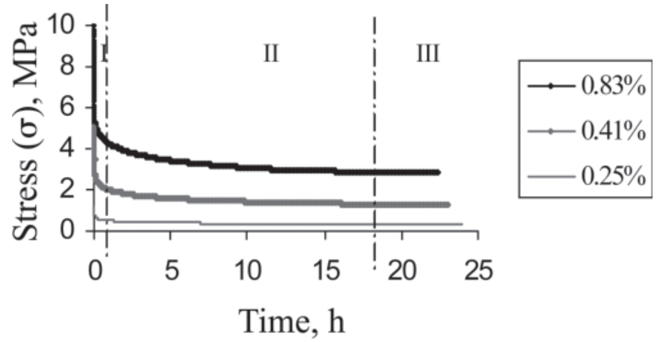


Fig. 8 Experimental compressive stress relaxation of HDPE versus time for different applied strain levels

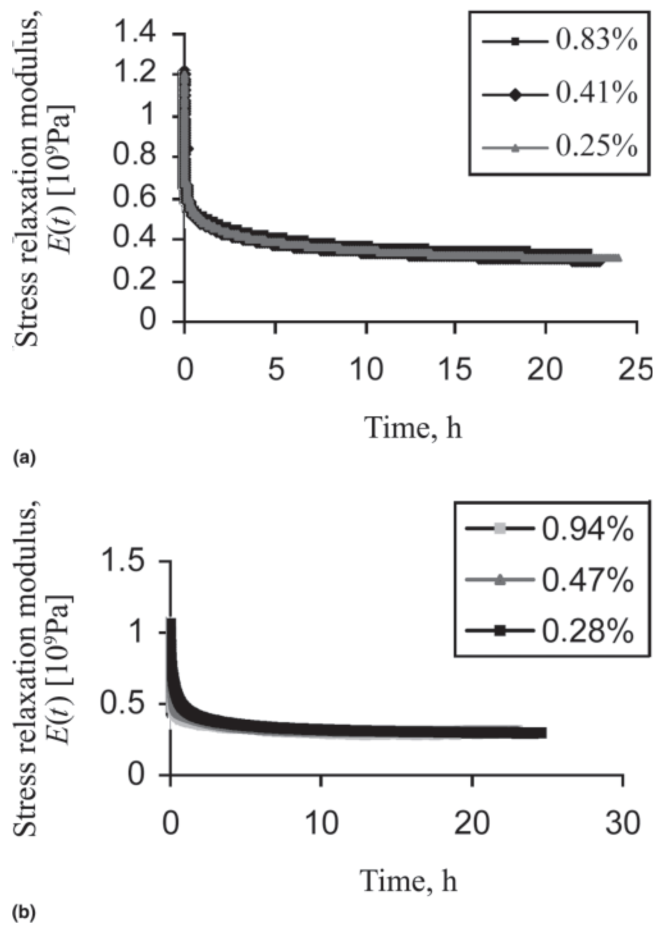


Fig. 9 (a) Experimental compressive stress relaxation modulus $E(t)$ of HDPE versus time for different applied strain levels; (b) experimental tensile stress relaxation modulus $E(t)$ of HDPE versus time for different applied strain levels

the linear domain of stress-strain curve. A viscoelastic linear behavior is observed for HDPE at low strain levels in the linear domain of monotonic compression test. This behavior has already been confirmed by creep tests.

4. Conclusions

The viscoelastic response of HDPE under tensile and compressive static long-term of creep was studied. Monotonic experiments under tensile and compressive tests were performed at different strain rates. In these experiments, the transition in the damage mechanisms was analyzed. The behavior of the HDPE was found to be nonlinear and dependent on the strain rates. The elastic modulus and plastic flow stress presented an increase with the displacement speed caused by the viscoelastic behavior of HDPE. In addition, the long-term behavior of HDPE was evidenced through creep tests at an imposed range of lower stresses and through stress relaxation tests at an imposed range of lower strains. It was found that the associated normalized curves could be represented by a single curve characterizing the compressive creep compliance or stress relaxation versus time. The viscoelastic behavior was confirmed to be linear within the linear domain of the monotonic compressive and tensile tests.

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