

Tensile Creep Behavior of Medium-Density Polyethylene

M. T. Hamed Mosavian* and A. Bakhtiari

Department of Chemical Engineering, Faculty of Engineering, Ferdowsi University of Mashhad, Azadi Square, Mashhad, Iran

S. Sahebian

Department of Materials Science and Engineering, Engineering Faculty, Ferdowsi University of Mashhad, Azadi Square, Mashhad, Iran

ABSTRACT: This research investigated the effect of stress and temperature on the viscous behavior of medium-density polyethylene (MDPE). The creep properties of pure MDPE using film specimens were studied under the static loading range 15–55 MPa by a thermomechanical analyzer at room temperature and elevated temperature (30°C, 60°C, and 90°C). Viscoelastic properties were derived from experimentally measured creep data, including isochronous stress–strain diagram, creep modulus, and creep rate charts with the influence of stress and temperature. In addition, the effect of creep strain and viscous flow on MDPE structure was investigated by a thermogravimetry analyzer. The results of this study showed that the linear viscoelastic behavior of MDPE changes with stress and temperature, and any reduction of polymer chains and structure revolution under the tensile creep test were not shown.

KEY WORDS: creep behavior, MDPE, viscoelasticity.

INTRODUCTION

THERMOPLASTIC POLYMERS, AS engineering materials, are widely used in many fields such as automotive and aviation industries, because of suitable mechanical and physical properties [1,2]. Low density, good

*Author to whom correspondence should be addressed.
E-mail: mosavian@um.ac.ir; hmosavian@gmail.com
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chemical and electrical receptivities, optical transparency, flexibility, etc., lead to suitable selection for many consumers. Nevertheless, low melting points and poor thermal properties have limited its application in industry. A main deficiency of thermoplastics is a lower dimensional stability exposed to a constant load for a period of time [3]. It means that the main deficiency of thermoplastics is the relatively poor creep resistance [4–8], which is a big barrier for their further expansion of industrial application.

Creep is a time-dependent deformation, which takes place under stress lower than the yielding strength of materials. Polymer and composite base polymer usually show long-term mechanical behavior under mechanical and/or environmental loadings due to viscoelastic behavior even at low temperature.

Material properties, such as material types [9], molecular orientation, density [10], crystallinity [11], etc., and external test conditions, such as applied load [12,13], temperature [14,15,11], and humidity caused to complicate the creep deformation of thermoplastic materials.

It is known that the relatively poor creep resistance of thermoplastic polymers is unfavorable to their application in industry. So, the viscous behavior of thermoplastics was the objective of extensive experimental studies over many decades [1–15]. Some researches can be summarized as follows:

Kolařík et al. investigated the nonlinear tensile creep of polypropylene (PP): time–strain super position and creep prediction. They showed that the compliance of PPs decreased with their crystallinity, while their creep rates are almost identical. Only rubber-toughened PP does show a slightly higher creep rate, which is attributed to the softening effect of rubber particles in the PP matrix [16]. Nonlinear creep behavior of viscoelastic polycarbonate was studied by Jazouli et al. The experimental and modeled results showed that the creep compliance above 26 MPa was stress-dependent and the material was nonlinear viscoelastic in nature [17].

The effect of stress and the type of material on the long-term creep behavior of pure polymers (PEN, polyethylene naphthalate; PET, polyethylene terephthalate; PBT, polybutylene terephthalate; PP; Nylon 6; and Nylon 66) and glass-reinforced thermoplastics were investigated by Lyons [9]. He showed that all types of samples had a linear viscoelastic behavior [9].

Medium-density polyethylene (MDPE) is one of the most important thermoplastic polymers. The main goal of this article is related to the effect of both stress and temperature on whole (linear and nonlinear) viscoelastic behavior of MDPE, and determination of polymer

structure due to creep strain over a short and long period of time of the creep test.

EXPERIMENTAL

Materials

MDPE was supplied by Tabriz Petrochemical Complex, Iran. The MDPE with density of 0.937 g/cm^3 , Vicat softening point of 117°C , and melting flow index (MFI) of $(2.16 \text{ kg at } 190^\circ\text{C}) \text{ g}/10 \text{ min}$ (4.2) was used as the beginning material. The crystallinity index of polyethylene is 62%. According to tension test using thermomechanical analyzer (TMA), it did not observe the fracture in the sample. The yield stress is 288 MPa. Strain to failure was not detected.

Specimen Preparation

To investigate the viscous behavior of MDPE, the films of MDPE were prepared by compression molding. For this purpose, the powders were compressed under 20–30 kPa at $160\text{--}180^\circ\text{C}$ for 5 min, followed by cooling to room temperature. The cooling rate is about $10\text{--}30^\circ\text{C}/\text{min}$. Due to cooling rate variation, the amount of fusion heat changes between 162 and 154 J/g , which is not a significant change.

Finally, the samples with final dimensions of $0.1 \times 0.1 \times 5 \text{ mm}^3$ were performed.

Thermomechanical Analyzer

Creep experimental data were recorded using TMA 60 in tensile mode. The samples with dimensions of $0.1 \times 0.1 \times 5 \text{ mm}^3$ were cut from the MDPE sheets. In order to elucidate the viscoelastic behavior of MDPE, the creep tests were carried out with various static forces of 15, 20, 30, 40, 45, and 55 MPa for 1 h. Moreover, to determine the influence of temperature, the creep behavior of MDPE at three different temperatures, that is, 30°C , 60°C , and 90°C , was measured with complete static forces. The results were reported as an average of the three tested samples.

Thermogravimetry Analysis

Thermogravimetry analysis (TGA) was done in a Shimadzu TGA 60. Samples of 2–3 mg of MDPE were heated from room temperature to

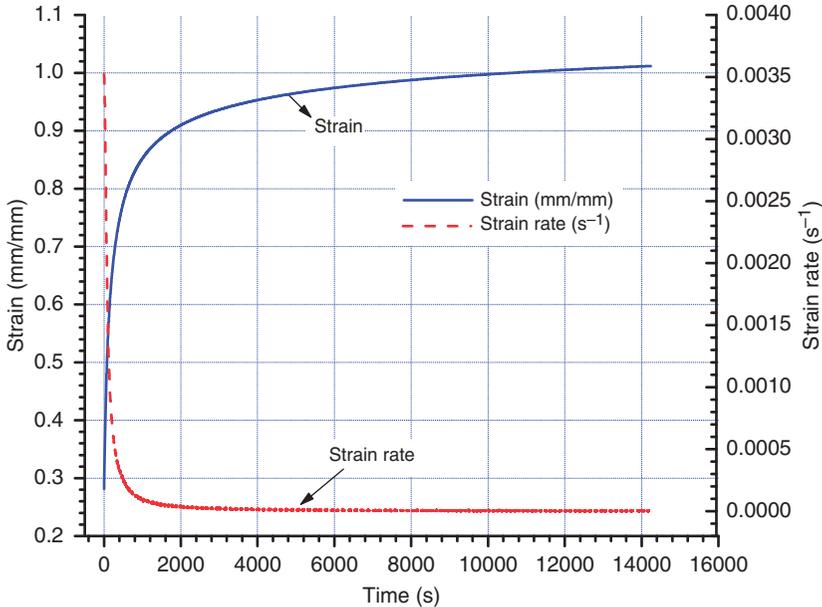


Figure 1. Creep strain and strain rate of MDPE vs time for creep stress of 15 MPa at room temperature.

600°C at a heating rate of 10°C/min. Averages of the three samples were reported.

RESULTS AND DISCUSSION

Creep Deformation

CREEP STRAIN

The creep strain and strain rate of MDPE versus time for creep stress of 15 MPa at room temperature are shown in Figure 1. Generally, a creep strain *versus* time could be divided into four stages, as shown in Figure 1. At first, initial rapid elongation would be shown at the beginning of the creep test that is clearly time-independent for all materials. Afterward, during the primary creep stage, the strain rate is relatively high, but it slows down with increasing strain, which may be due to the slip page and re-orientation of polymer chains under persistent stress. After a certain period, the strain rate eventually reaches a minimum and nearly becomes constant; this is known as secondary or steady-state creep. At this stage,

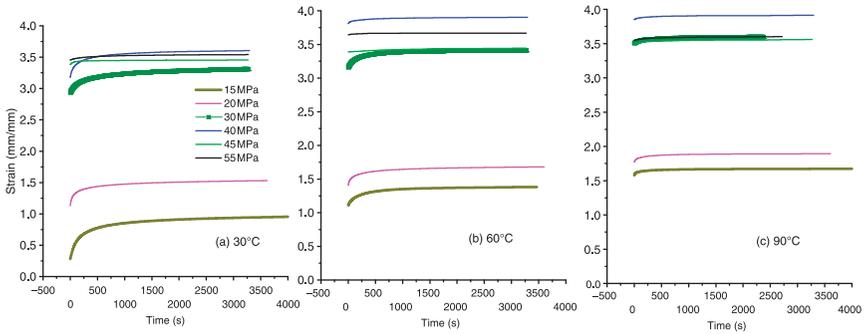


Figure 2. The curves of creep strain vs creep time of MDPE under various static loads and temperatures.

viscous flow activates, and a dynamic equilibrium of polymer structure evolution and external load is reached subsequently. The duration of this stage is relatively long. Finally, at the fourth stage, known as tertiary creep stage, the creep rate increases rapidly and the final fracture occurs.

According to Figure 1, initial rapid elongation, the primary creep, and unfinished secondary creep stages after 4 h, were shown for MDPE samples under creep stress of 15 MPa. The curves of creep strain *versus* creep time of MDPE under different static loads (15, 20, 30, 40, 45, and 55 MPa) and different temperatures (30°C, 60°C, and 90°C) were shown in Figure 2. Based on Figure 2, the creep stages were clearly shown in the figures as instantaneous deformation, primary, and unfinished second creep stages.

The creep deformation at three stages of MDPE samples increased as a rising temperature. The incremental creep strain of MDPE samples by increase in temperature with a stress lower than 30 MPa was significantly represented, while the creep strain at a higher stress of around 30 MPa was almost the same. The reason of this effect was related to stress activation of polymer chains, so they did not act as a rising temperature. Besides, an increase in stress up to 30 MPa, as shown in Figure 2, resulted in an extensively increasing trend of creep deformation, while at a stress higher than 30 MPa, the creep deformation represented a limited incremental trend. The reason of this effect probably could be attributed to similar work hardening and re-orientation hardening of polymer chains under the existent load.

INITIAL RAPID CREEP STRAIN

Another interesting result of the creep experimental data was the initial rapid creep strain. Figure 2 shows the effect of temperature and stress on

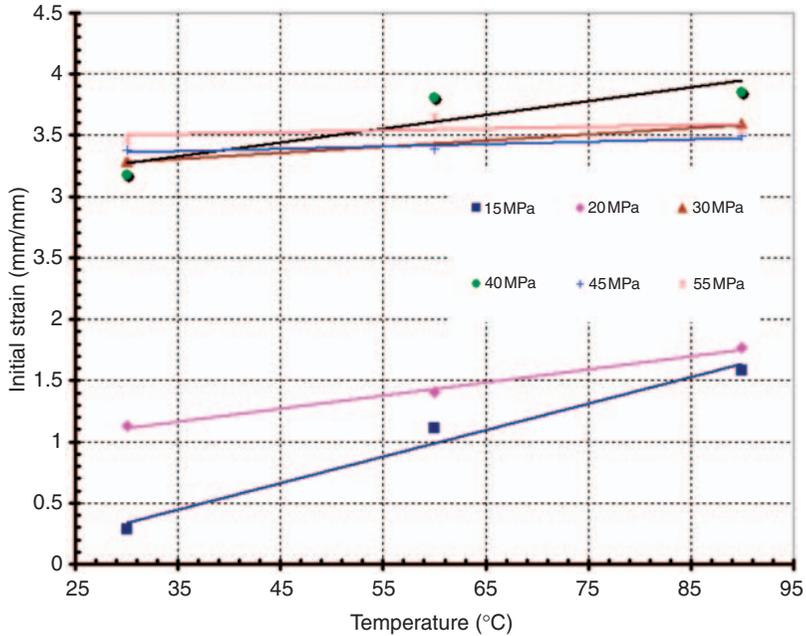


Figure 3. The effect of temperature and stress on initial creep deformations of MDPE samples.

initial creep deformations of MDPE samples. As shown, instantaneous (plastic and elastic) deformation of MDPE samples considerably increased at stress lower than 30 MPa, while initial rapid creep strain had almost the same at lower stress than 55 MPa and higher than 30 MPa. The reason for this effect is related to the similar amount of MDPE chains' orientation along existent stress.

Also, increasing mobility chains of polymer caused an incremental creep deformation due to rising temperature. According to Figure 3, MDPE chains reacted to the temperature at stress lower than 30 MPa, so the variation of temperature significantly affected creep deformation. When the applied load was much more than 30 MPa, polymer chains had a stress-activated behavior. Therefore, creep deformation had a lower slope versus temperature with respect to MDPE samples at lower stress than 30 MPa.

ISOCHRONOUS STRESS-STRAIN DIAGRAMS

Additional information about material response to time could be achieved by cross-plotting the experimental data in the form of isochronous

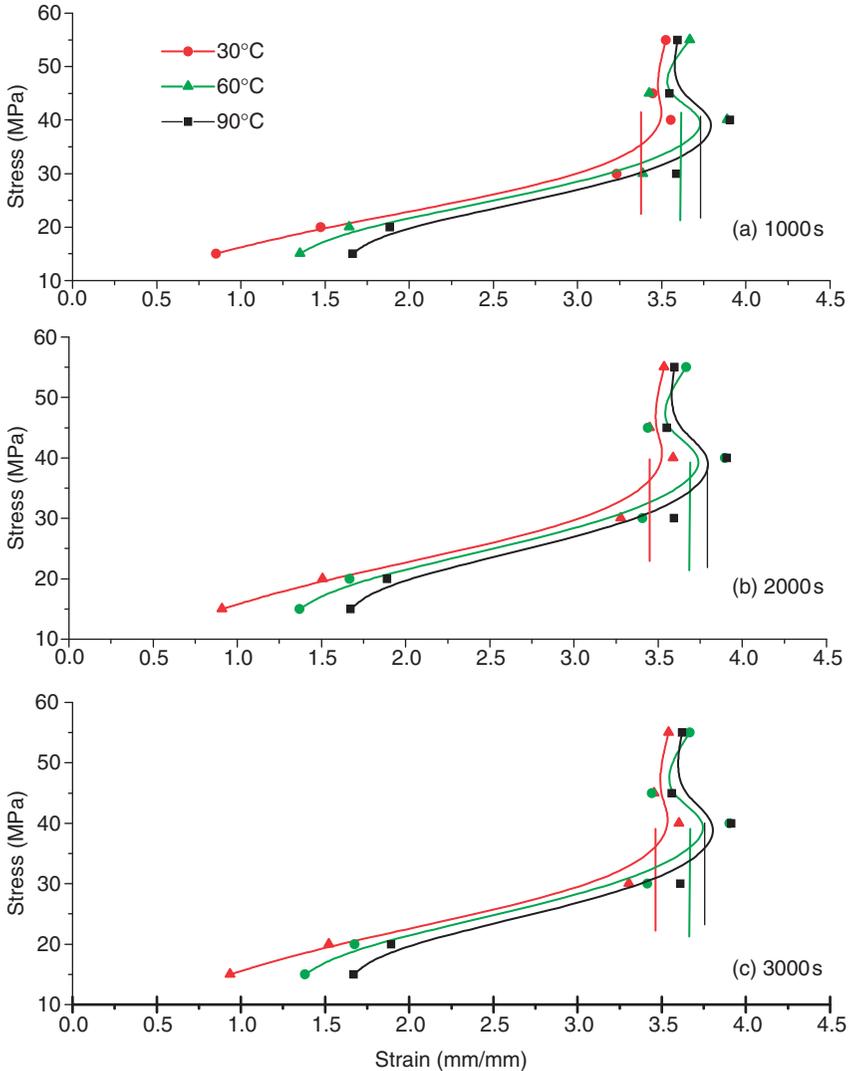


Figure 4. Isochronous stress–strain diagrams for MDPE for times of 1000, 2000, and 3000 s.

stress–strain diagrams. A curve of the applied stress versus the strain at a given time is an isochronous diagram. In this article, the experimental data (stress–strain) at 1000, 2000, and 3000 s were used to produce the isochronous stress–strain diagrams. The results are shown in Figure 4. It could be realized from this figure that a linear relationship at stress lower than 30 MPa and higher than 15 MPa, was shown for MDPE samples. Also,

at an elevated temperature, the creep deformation of MDPE samples increased, as compared to room temperature under the same stress level.

Looking at Figures 3 and 4 in more detail, it might be concluded that linear viscoelasticity was observed at the stress level between 15 and 30 MPa, so polyethylene chains did not act as an existent stress, and creep deformation was significantly affected by temperature. An important note is shown in Figure 4. The linear domain of stress-strain was affected by temperature, and the linear relationship of stress-to-strain domain increased by rising temperature. Besides, at strain 350% or at a stress level higher than 30 MPa, the creep behavior of MDPE changes. The linear viscoelasticity of MDPE transforms to nonlinear viscoelasticity due to high stress level.

Retardant Viscous Time

One of the main interesting results of the experimental creep data was the retardant viscous time known as the duration of primary creep stage. Temperature and stress affected the duration of primary creep stage. Based on Table 1, both stress and temperature cause an increase in the viscous behavior of MDPE, and lower the retardant viscous time.

The retardant viscous time depends on the reaction of polymer chains, including re-align and re-orientation of MDPE chains. According to dependency of retardant viscous time and polymer chain reaction, both temperature and stress affect on polymer chain reaction and it occurs in a lesser time by an increase in temperature and stress. According to Table 1, the linear and nonlinear viscoelastic behaviors of MDPE did not affect retardant viscous time. So, the reduction trend of time by increase in temperature and stress was shown.

Table 1. Retardant viscous time of MDPE under various static loads and temperatures.

		Retardant viscous time (s)			Retardant viscous time (s)			Retardant viscous time (s)
15 MPa	30°C	3334	30 MPa	30°C	2500	45 MPa	30°C	1600
	60°C	2500		60°C	1950		60°C	1550
	90°C	1500		90°C	1360		90°C	1180
20 MPa	30°C	2700	40 MPa	30°C	2100	55 MPa	30°C	1400
	60°C	2180		60°C	1700		60°C	1350
	90°C	1400		90°C	1300		90°C	1150

Creep Strain

The dimensional stability of materials was determined by another important parameter, that is, creep rate, which represented the velocity of creep deformation. The creep rate of MDPE samples is shown in Figure 5 under different stress levels and temperatures. Based on this figure, in the linear viscoelasticity domain, the creep rate of MDPE samples decreased with increasing temperature at stress levels lower than 30 MPa. The reason for this effect was related to thermally activated polymer chains. In fact, at elevated temperatures, the polymer chains were thermally activated and easily deformed, so some polymer segments re-aligned to stress direction and orientational hardening of MDPE increased. As a result, polymer chains did not undergo further re-orientation and re-arrangement due to relatively low stress [15].

As shown in Figure 5, at stress level higher than 30 MPa and lower than 55 MPa, the creep rate of each sample increased with increasing temperature as a nonlinear viscoelastic behavior. This observation could be attributed to stress activated by MDPE chains. The segments of MDPE chains activated at a high stress level. Although a considerable instantaneous deformation was shown due to the high value temperature and stress, re-orientation and re-arrangement of polymer chains increased. But high stress level activated polymer chains, and the incremental creep rate was shown by rising temperature.

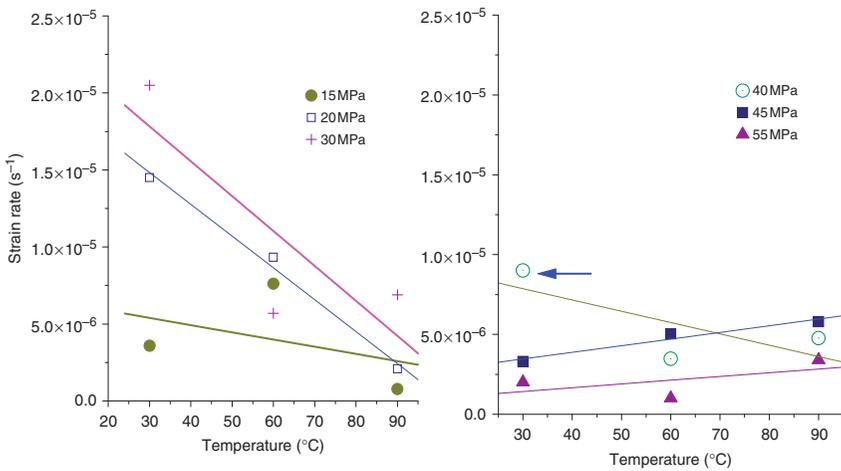


Figure 5. The secondary creep rates of MDPE at different temperatures and stress levels.

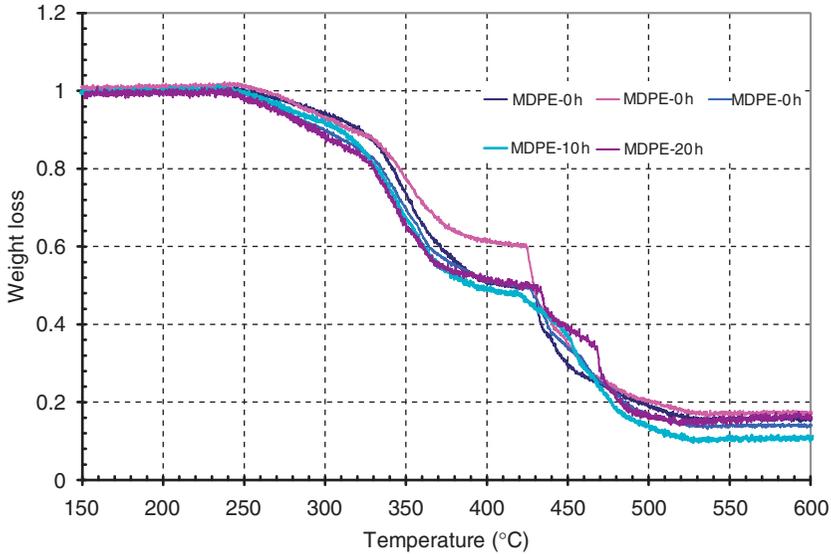


Figure 6. The weight loss of MDPE vs temperature at different creep times.

Thermogravimetry Analysis

In order to investigate the viscous flow mechanism on polymer structure, the stress whitening zone of MDPE specimens after creep test was cut and thermogravimetry test under low tensile creep stress was done. One of the main mechanisms in creep test was the reduction of polymer chains. Figure 6 shows the weight loss of MDPE versus temperature. As analyzed above, after 20 h of the creep test of MDPE, the weight loss of MDPE samples was similar to MDPE samples, after 1 h of the creep test. It seems that below 20 h of creep test under low stress, the reduction of MDPE chains did not occur significantly.

CONCLUSIONS

In this study, MDPE films were made and were investigated using a thermomechanical analyzer test and a thermogravimetry analyzer. The results were summarized as follows:

1. Creep rates and instantaneous deformation of samples increased by rising temperature and stress levels, but the dependence of viscous deformation of MDPE was related to the reaction of polymer segments to stress and temperature.

2. Creep rate under a low stress level, lowered by rising temperature due to the thermally activated MDPE chain segments.
3. At a high stress level, the segment of polymer chains was activated with respect to stress, and the creep rate had a completely incremental trend by increase in temperature.
4. The linear and nonlinear viscoelastic domains were affected by variation of temperature.
5. At a low stress level, the deterioration of polymer did not occur after a long and short creep time.
6. The duration of the primary creep stage changes with temperature and stress applied to materials. Rising stress and temperature decreased retardant viscous time.

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