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Time and temperature dependent response and relaxation of a soft polymer

Akhtar S. Khan*, Oscar Lopez-Pamies¹

*Department of Mechanical Engineering, University of Maryland Baltimore County,
Baltimore, MD 21250, USA*

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Abstract

A comprehensive study on the compressive uniaxial mechanical response of the polymer Adiprene-L100, a polyether urethane-based rubber, is presented. Quasi-static and dynamic experiments, under compression, were performed at a wide range of strain rates ($10^{-5} \leq \dot{\epsilon} \leq 5000 \text{ s}^{-1}$) and temperatures ($-100 \leq T \leq 270 \text{ }^\circ\text{F}$). The behavior of the polymer was found to be nonlinear and highly dependent on both, strain rate and temperature. Adiprene-L100 exhibited stress relaxation, dependent on strain rate, initial strain and stress levels at room temperature. The viscoplastic strain was found to be negligibly small. © 2002 Published by Elsevier Science Ltd.

Keywords: Stress relaxation; Polymeric materials

1. Introduction

There has been an increased use of polymeric materials in structures such as binder-constituents in explosives, load-bearing components, automobiles, jet engines, etc. in recent years. Experimental data for simple and complex loading paths are increasingly needed with the growing use of polymers in various fields (Bardenhagen et al., 1997). According to previous studies, detailed knowledge of the effects of temperature and strain rate as well as stress relaxation and creep processes on polymers is essential for developing a good understanding of their mechanical

* Corresponding author. Tel.: +1-410-455-3301; fax: +1-410-455-1052.

E-mail address: khan@umbc.edu (A.S. Khan).

¹ Now at the Department of Mechanical Engineering and Applied Mechanics, The University of Pennsylvania.

behavior (Khan and Zhang, 2001; Krempl, 2001). Gray et al. (1997) performed experiments at three strain rates (0.001, 0.1, 3000 s⁻¹) at room temperature, and at six temperatures at a strain rate of 3000 s⁻¹ on Adiprene-L100. This study seemed preliminary in nature, especially regarding the dependence on strain rate, as they found almost no dependence at the two lower strain rates, but a large jump in the response at the highest strain rate. The aim of the present paper is to provide a detailed study of the compressive uniaxial mechanical behavior of the polymer Adiprene-L100 under a wide range of temperatures ($-100 \leq T \leq 270$ °F) at $\dot{\epsilon} = 0.1$ s⁻¹, and strain rates ($10^{-5} \leq \dot{\epsilon} \leq 5000$ s⁻¹) at room temperature as well as the stress relaxation response in the finite strain regime. Note that the strain rate $\dot{\epsilon}$ refers to approximate true strain rate. The results from this investigation will be critical to constitutive modelers with different perspectives. Multiaxial experiments (to be presented in a different paper) such as non-proportional biaxial compression, pure torsion, tension–torsion and torsion–tension at different strain rates will also be performed on Adiprene-L100. These multiaxial experiments in addition to the uniaxial experimental study presented in this paper will provide the comprehensive response of the polymer Adiprene-L100 in the finite deformation regime. Most of the literature on polymers reviewed by the authors was limited to narrow regimes of strain, strain rate, and temperature (McGuirt and Lianis, 1970).

2. Experimental procedures

2.1. Material

All the specimens utilized in this study were obtained from an Adiprene-L100 formed in a plate or slab. Adiprene-L100 is a polyether urethane-based rubber that has outstanding abrasion resistance and excellent hydrolytic stability. It also has excellent dynamic properties, such as flex resistance and low heat build-up. It reaches a maximum elongation of 560% at the breaking point. The glass transition temperature is $T_G = -80$ °F (-62 °C).

2.2. Quasi-static experiments

Solid cylinders of Adiprene-L100 with a length of 1.00 in (25.4 mm) and a diameter of 0.80 in (20.5 mm) were used for the compressive quasi-static, temperature and time dependent experiments. All of the quasi-static experiments were performed under displacement control on a MTS axial/torsional servo-hydraulic 810 system. The specimens were compressed between platens made of VascoMax C-35 steel which was heat-treated to its maximum hardness. The displacement data given by the MTS machine were utilized to determine the strain in the specimens. No correction was necessary given that the deformation of the machine during the loading was negligible. In order to reduce the friction between the interfaces of the specimens with the platens and to avoid barreling, different lubricants (as specified below) were used.

2.2.1. Temperature dependent experiments

Uniaxial compressive tests at strain rate of 10^{-1} s^{-1} with constant temperatures of -100 , 20 , 70 , 185 , and 270 °F (-73 , -7 , 21 , 85 , and 132 °C) were performed. Dow Corning® 33 low temperature lubricant was applied on the bases of the -100 °F (-73 °C) specimen in order to maintain a uniform stress state. This temperature was reached and stabilized by submitting the specimen to a dry ice bath 6 h prior to and during the test. The same low temperature lubricant was utilized for the 20 °F (-7 °C) experiment; the temperature for this test was stabilized in a calcium chloride solution with a melting temperature of 20 °F (-7 °C). Dow Corning® 41 high temperature lubricant was used on the 185 and 270 °F (85 and 132 °C) specimens in order to maintain a uniform stress state within them. These temperatures were reached and stabilized by using a heating tape six hours prior to and during the test. The standard vacuum grease was used as the lubrication for the experiment performed at 70 °F (21 °C). A digital thermometer (Omega 871A) was used to monitor the temperature during the stabilization and loading processes.

2.2.2. Time dependent experiments

Standard vacuum grease was used on all the time dependent experiment specimens in order to achieve a uniform stress state. Uniaxial compressive tests at room temperature of $T=70$ °F (21 °C) with constant strain rates of 10^{-5} , 10^{-4} , 10^{-2} and 10^0 s^{-1} were performed up to a true strain level of 55% and unloaded. Furthermore, other compression specimens were taken to approximate true strain levels of $\varepsilon=5$, 10, 25, and 55% under loading constant strain rates of 10^{-4} , 10^{-2} , and 10^0 s^{-1} and unloaded. Uniaxial compressive single stress relaxation experiments at room temperature were performed at loading and unloading strain rates of 10^{-4} , 10^{-2} , and 10^0 s^{-1} . The specimens were loaded approximately up to $\varepsilon=60\%$ where the displacement was held constant for 2 h. After the holding period, the specimens were unloaded to zero stress. Uniaxial compressive multiple stress relaxation experiments were performed under loading and unloading strain rates of 10^{-4} , 10^{-2} , and 10^0 s^{-1} . During these tests, the specimens were loaded up to $\varepsilon=5\%$ where the displacement was held constant for 2 h. Then the specimens were loaded up to $\varepsilon=10\%$ where the displacement was held constant again for 2 h. This loading/holding process was repeated for true strain levels of $\varepsilon=5$, 10, 15, 20, 25, 30, 35, 40, 45, and 50%.

2.3. Dynamic experiments

Dynamic compression experiments with an approximately constant strain rate of 5000 s^{-1} were performed using the Split-Hopkinson Pressure Bar technique (SHPB). The pressure bars were made of 0.50 inch (12.7 mm) diameter Al 2024-T351 rods. The specimens were solid disks of Adiprene-L100 of 0.42 inch (10.67 mm) diameter and 0.13 inch (3.30 mm) thickness. The detail description of the SHPB technique is given in Liang and Khan (1999), Khan and Liang (1999), Khan and Zhang (2000).

3. Experimental results and discussion

All the quasi-static and dynamic stress and strain results of the uniaxial mechanical behavior of the polymer Adiprene-L100 are based on the current configuration of the specimen; i.e. true stress and true strain are utilized. Recall that true strain is given by

$$\varepsilon = \ln(\lambda) \tag{1}$$

where λ is the stretch ratio in the loading direction, which is easily determined from the MTS displacement data. True stress is given by

$$\sigma = \frac{P}{A} \tag{2}$$

where P is the applied load, given by the MTS, and A is the current (deformed) cross sectional area of the specimen. If the material is incompressible Eq. (2) can be rearranged in a more convenient way

$$\sigma = \frac{P}{A_0} \cdot \lambda \tag{3}$$

where A_0 is the initial cross sectional area. Eq. (3) is derived under the assumption that the material undergoes no volume change during loading. This is not necessarily true for polymers. Preliminary studies on compressive loading at room temperature and strain rate of 10^{-4} s^{-1} were done to up to true strain levels of 50% in

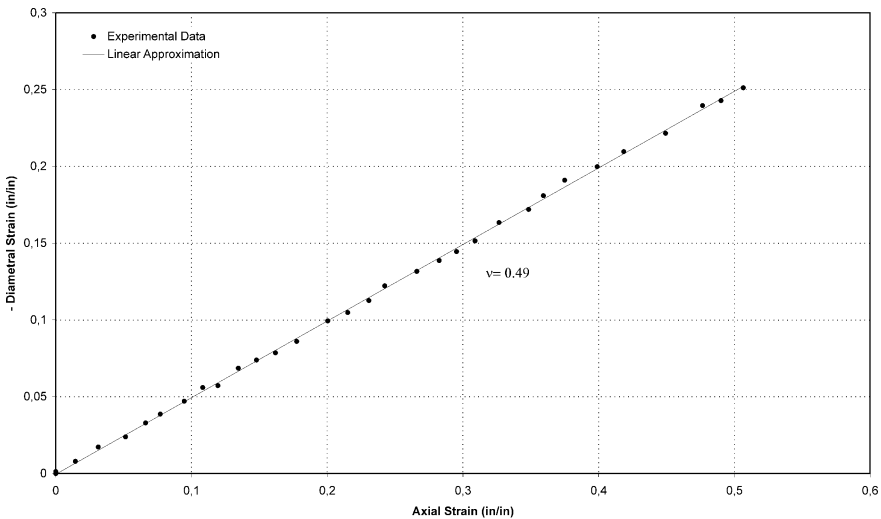


Fig. 1. Poisson ratio of Adiprene-L100 determined from uniaxial compression experiments.

order to investigate if uniaxial compression was an isochoric process on Adiprene-L100. Both, the diametral and the axial strain were monitored during these experiments. Fig. 1 shows the negative true diametral strain versus the axial strain (compressive strains are assumed positive). An approximate constant Poisson ratio of $\nu=0.49$ was obtained. Furthermore, Fig. 2 shows the change in volume per unit volume as a function of true axial strain. Based on these figures, it was concluded that Adiprene-L100 undergoes very small (negligible) volume contraction during compression and the use of true stress is consequently justified. A major concern during this uniaxial experimental study on Adiprene-L100 was the reproducibility of results, given the characteristic low strength of the material. Numerous specimens were utilized for the same specific experiment. The results were satisfactory, as they indeed were reproducible. Fig. 5 shows the stress–strain relationship for constant strain rate of 10^{-4} s^{-1} up to different strain levels of 5, 10, 25 and 55% using four different specimens. Clearly the behavior of the four specimens was very similar in the pertinent regions. This pattern was observed throughout the presented experimental study on Adiprene-L100. The maximum deviation in stress noted from specimen to specimen for any given experiment was 4%.

3.1. Temperature dependent experiments

Fig. 3 shows the stress–strain relationship for the different temperature tests performed on Adiprene-L100 at strain rate of 10^{-1} s^{-1} . The material response at different temperatures changes significantly. The glass transition temperature effect appears very clearly in this figure. For Adiprene-L100, this phenomenon occurs at $T_G = -80 \text{ }^\circ\text{F}$ ($-62 \text{ }^\circ\text{C}$), below this temperature, the initial modulus drops by several

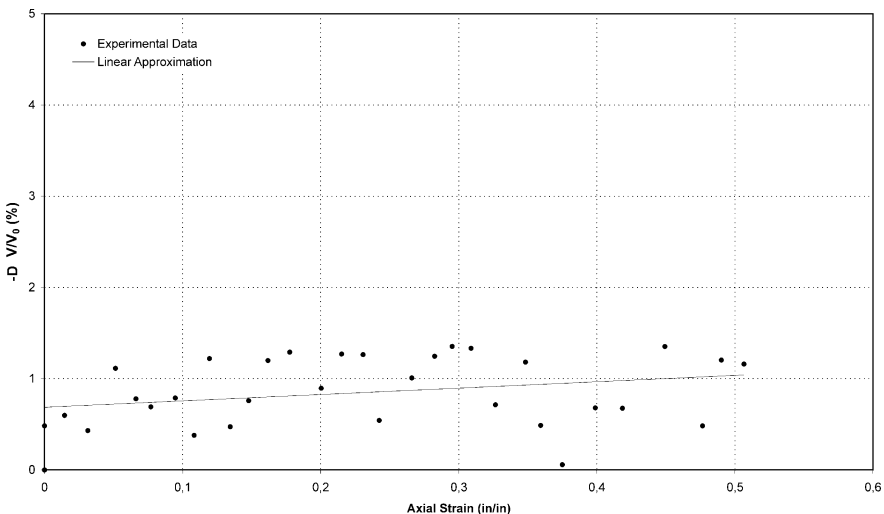


Fig. 2. Change of volume per unit volume versus axial strain for Adiprene-L100 determined from uniaxial compression experiments.

orders of magnitude providing a transition from “glassy” to “rubbery”. The compressive response of Adiprene-L100 at different temperatures is nonlinear during loading. There exist, though, two almost linear regions, namely the initial response or initial modulus and the asymptotic behavior at large strains ($\epsilon > 10\%$). The dependence on temperature increases nonlinearly with the decrease of temperature. Also, the deformation of Adiprene-L100 is mainly elastic but there exist some residual strain upon unloading. The geometry of the specimens was monitored for several days after unloading (with calipers). The specimens underwent some recovery in strain during this period but there is a small temperature-dependent viscoplastic deformation. The largest viscoplastic strain observed was approximately 3% at the highest temperature experiment, i.e. $T = 270^\circ\text{F}$ (132°C). A detailed characterization of the viscoplastic strain undergone by Adiprene-L100 is given below.

3.2. Time dependent experiments

Fig. 4 shows the loading stress–strain relationship for the different strain rates of $\dot{\epsilon} = 10^{-5}$, 10^{-4} , 10^{-2} , 10^0 , and 5000 s^{-1} tests performed on Adiprene-L100 at room temperature 70°F (21°C). The nonlinear dependence of the compressive response of Adiprene-L100 on strain rate is clear. As in the previous case, there exist two almost linear regions, namely the initial response (or initial modulus) and the asymptotic behavior at large strains ($\epsilon > 10\%$). The dependence on strain rate is very significant and it increases nonlinearly with strain and strain rate. Similar to the glass transition temperature effect, Adiprene-L100 exhibited a drastic change, from “rubbery” to “glassy” behavior, between the strain rates of 5000 and 1 per second. This behavior is not characteristic of all polymers, unlike the glass transition effect (Lockett, 1972; Yuan and Khan, 2000). The behavior shown in Fig. 4 differs from the one found by

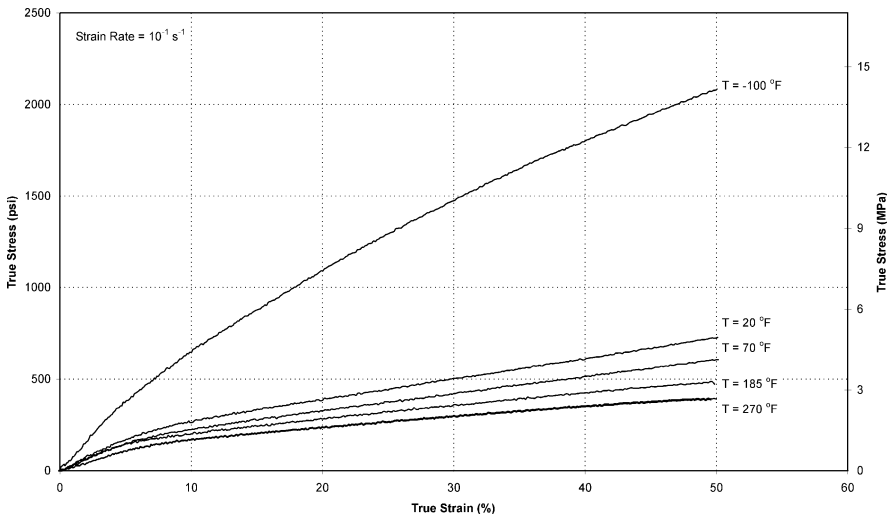


Fig. 3. Uniaxial compression experiments on Adiprene-L100 at different temperatures.

Gray et al. (1997) where the dependence of Adiprene-L100 under quasi-static strain rates was shown to be insignificant. It was found, as in the constant temperature experiments, that the deformation on Adiprene-L100 is mainly viscoelastic but there is small viscoplastic deformation. The specimens for different strain rate experiments were monitored for several days after unloading. A viscoelastic recovery was observed as well as permanent deformation, given that the specimens never recovered fully to their initial length. Both strains, viscoelastic and viscoplastic, were found to be strain rate sensitive. In order to find the specific dependency between strain rate and the

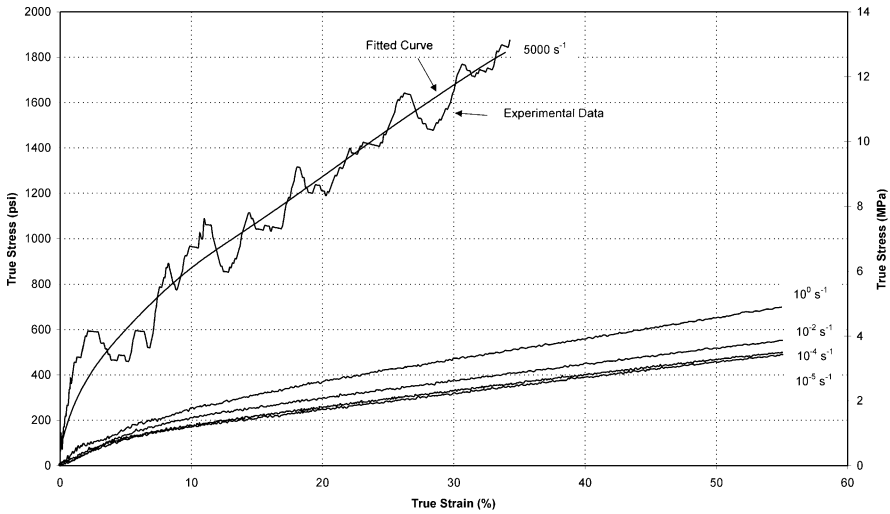


Fig. 4. Uniaxial compression experiments on Adiprene-L100 at different strain rates.

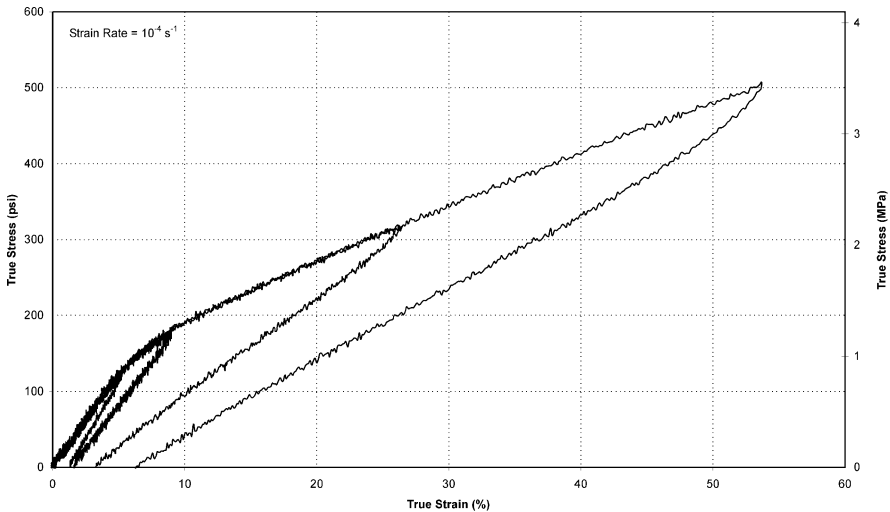


Fig. 5. Uniaxial compression experiments on Adiprene-L100 at different strain levels.

viscoelastic and viscoplastic strains, experiments up to different strain levels at different strain rates (specified above) were performed. Figs. 5–7 show the loading/unloading stress–strain relationship for approximate strain end levels of $\epsilon = 5, 10, 25,$ and 55% at strain rates of $\dot{\epsilon} = 10^{-4}, 10^{-2},$ and 10^0 s^{-1} , respectively. It was noted that the unloading is quasi-linear. The geometry of the specimens involved in these experiments was monitored (with calipers) beyond unloading until a stable configuration was observed. By knowing the maximum strain achieved by the specimens as well as the residual strain, a clear division between viscoelastic and viscoplastic

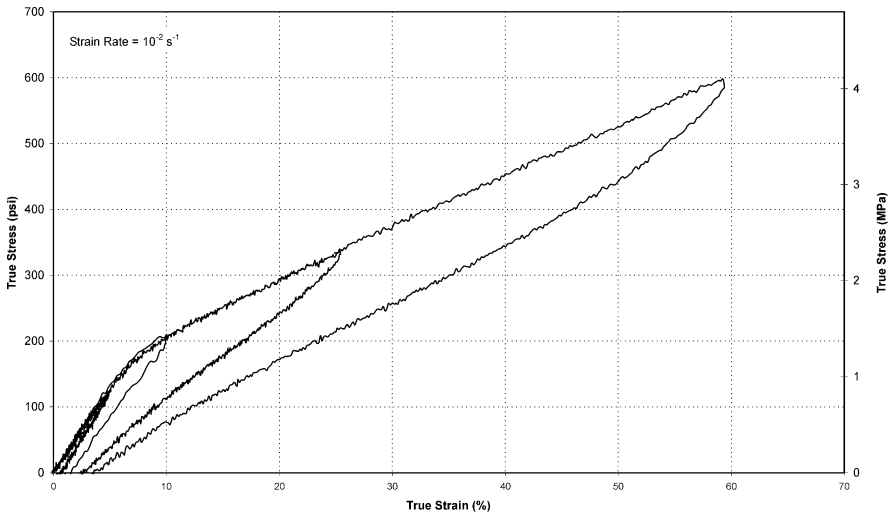


Fig. 6. Uniaxial compression experiments on Adiprene-L100 at different strain levels.

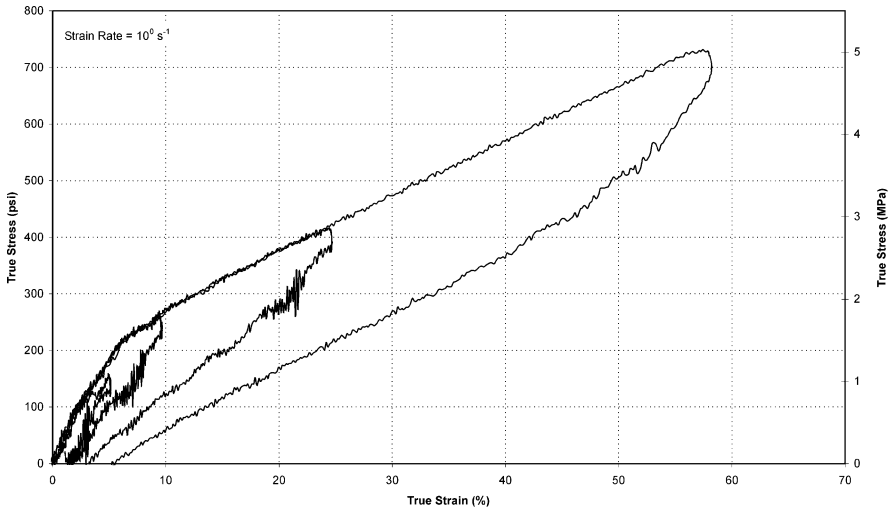


Fig. 7. Uniaxial compression experiments on Adiprene-L100 at different strain levels.

strains could be discerned. Fig. 8 shows the viscoplastic strain; i.e. permanent strain, as a function of the total true strain. The viscoplastic strain in Adiprene-L100 exhibited a strong dependency on strain rate; this is not the case for other polymers such as Teflon (Khan and Zhang, 2001). This dependence increases nonlinearly with decreasing strain rate. Fig. 9 shows the viscoelastic strain as a function of the total true strain. The dependency of viscoelastic strain on strain rate is small but significant. In order to acquire a quantitative notion of how much viscoplastic

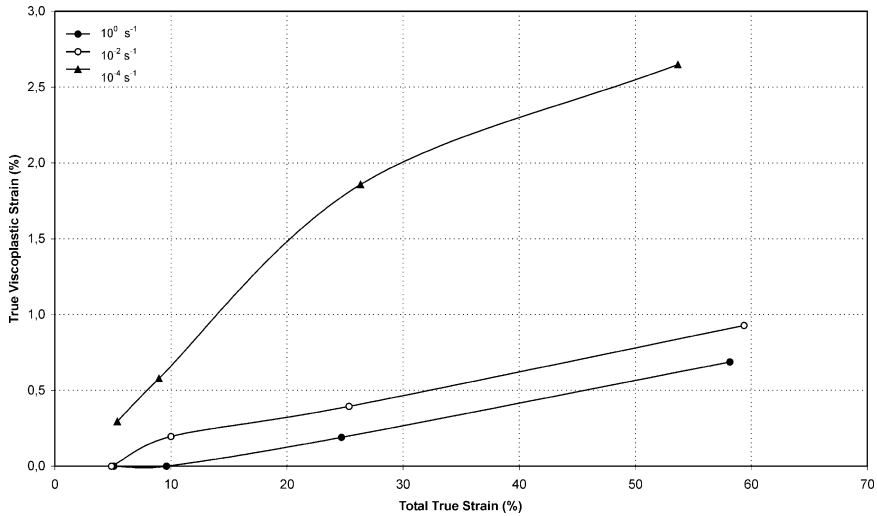


Fig. 8. Viscoplastic strain (Adiprene-L100).

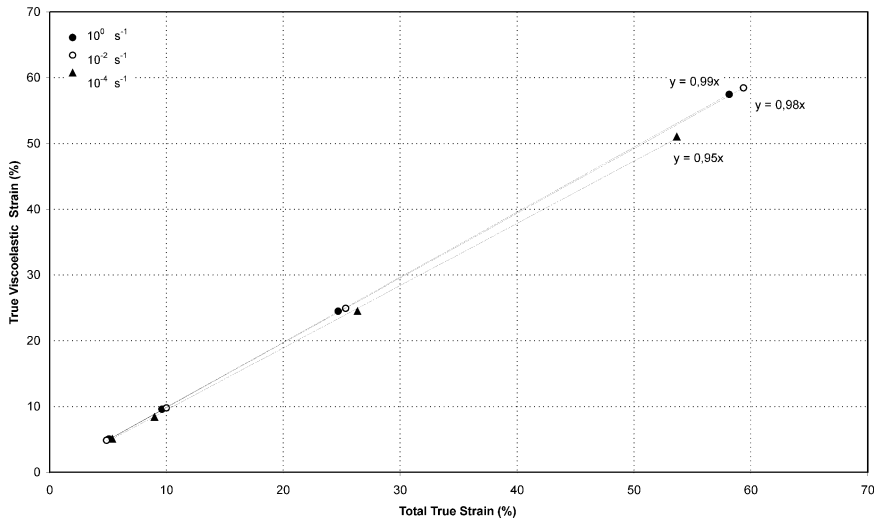


Fig. 9. Viscoplastic strain (Adiprene-L100).

deformation Adiprene-L100 undergoes, the viscoelastic strain of Fig. 9 was approximated by a linear behavior. Hence, according to the linear approximation from Fig. 9, only 5, 2, and 1% of the total strain is permanent for strain rates of $\dot{\epsilon} = 10^{-4}$, 10^{-2} , 10^0 s^{-1} , respectively. This clearly indicates that most of the deformation that Adiprene-L100 undergoes is viscoelastic. Fig. 10 shows the loading/unloading stress–strain relationship under constant strain rates of $\dot{\epsilon} = 10^{-4}$, 10^{-2} , 10^0 s^{-1} at room temperature ($T = 70 \text{ }^\circ\text{F}$, $21 \text{ }^\circ\text{C}$) with a single stress relaxation process around $\epsilon = 60\%$. As expected, Adiprene-L100 relaxed at a temperature as low as $T = 70 \text{ }^\circ\text{F}$ ($21 \text{ }^\circ\text{C}$). The differences in the relaxation behaviors at the different loading strain rates of $\dot{\epsilon} = 10^{-4}$, 10^{-2} , 10^0 s^{-1} are due to strain rate sensitivity and initial strain and stress levels. Fig. 11 shows how the reduction of the stress during relaxation from Fig. 10 with respect to time. It is clear how an increase in strain rate results in an increase in the amount of relaxation. Also, no constant equilibrium state was reached by Adiprene-L100 after 2 h of relaxation as it exhibited a slight asymptotic negative slope. Also, no apparent tendency to a common equilibrium state was exhibited by the polymer, as the end point of the relaxation processes was slightly different for every strain rate. Fig. 12 clearly shows how the end points of the single relaxation processes were lower the higher the strain rate. Despite this observed definite pattern, it should be noted that a maximum deviation of 4% has been monitored among the response of different specimens. The difference between the end points of the single step stress relaxation processes are around this percentage. Figs. 13–15 show the loading stress–strain relationship under constant strain rates of $\dot{\epsilon} = 10^{-4}$, 10^{-2} , and 10^0 s^{-1} at room temperature ($T = 70 \text{ }^\circ\text{F}$, $21 \text{ }^\circ\text{C}$) with multiple stress relaxation processes around $\epsilon = 5, 10, 15, 20, 25, 30, 35, 40, 45,$ and 50% , respectively. These figures also include the corresponding constant strain rate behavior of the polymer for comparison purposes. For the two lower strain rates, it

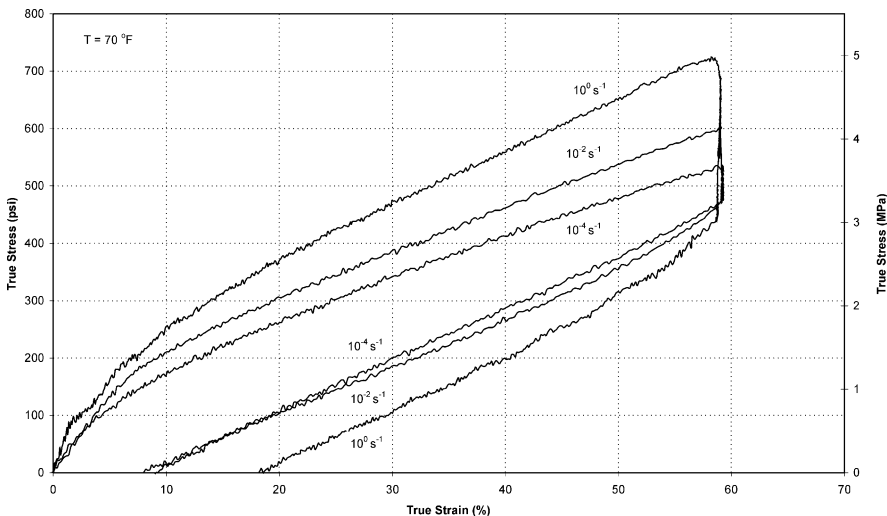


Fig. 10. Single step stress relaxation experiments on Adiprene-L100.

appears that the mechanical behavior of the polymer Adiprene-L100 is not altered by relaxation processes. The multiple step stress relaxation curves followed the constant strain rate behavior. This is not the case for the experiment at strain rate $\dot{\epsilon} = 10^0 \text{ s}^{-1}$. Fig. 15 shows how the constant strain rate behavior was stiffer than the relaxation one beyond the strain of $\epsilon = 10\%$. In synthesis, the multiple step stress relaxation behavior at $\dot{\epsilon} = 10^0 \text{ s}^{-1}$ followed the constant strain rate response up to

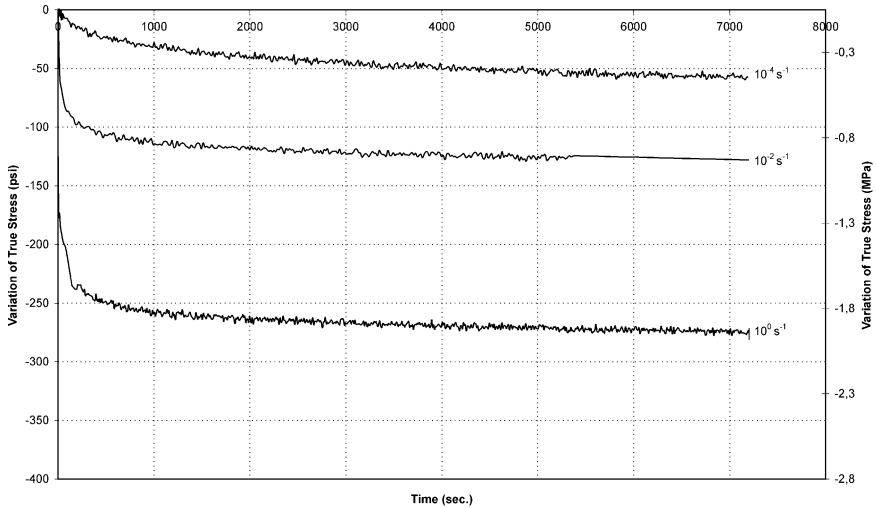


Fig. 11. Single step stress relaxation experiments on Adiprene-L100.

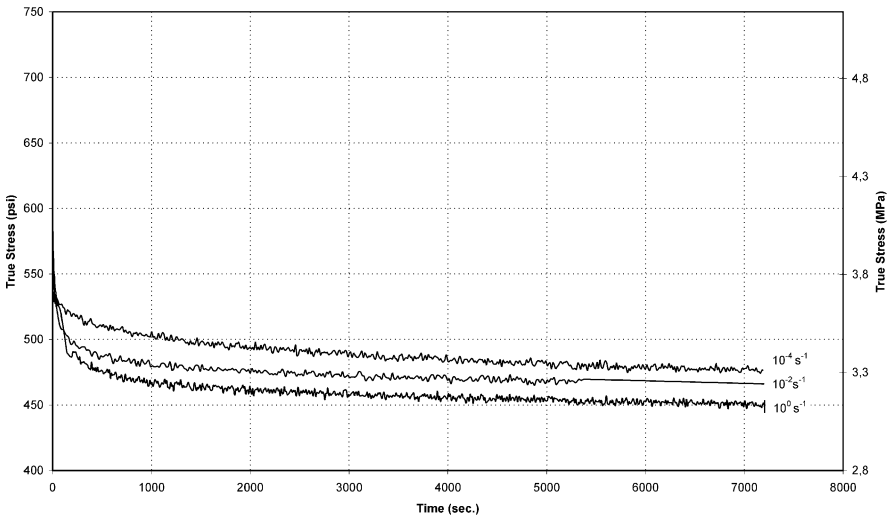


Fig. 12. Single step stress relaxation experiments on Adiprene-L100.

$\varepsilon = 10\%$ and the multiple step stress relaxation behavior at lower strain rates of $\dot{\varepsilon} = 10^{-4}, 10^{-2} \text{ s}^{-1}$ followed the constant strain rate response for the entire spectrum of the experiment. This indicates that relaxation processes do not alter the internal structure of the polymer Adiprene-L100. A likely explanation for the deviation from this characteristic in the case of the $\dot{\varepsilon} = 10^0 \text{ s}^{-1}$ test would be that the strain intervals at which the relaxation processes were performed were too small in the sense that upon reloading from relaxation, the material did not have enough time to reach the

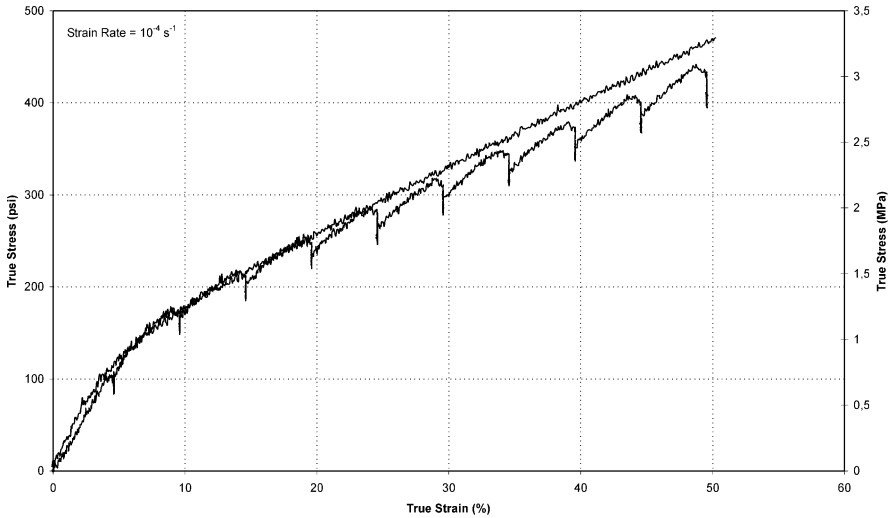


Fig. 13. Multiple step stress relaxation experiments on Adiprene-L100.

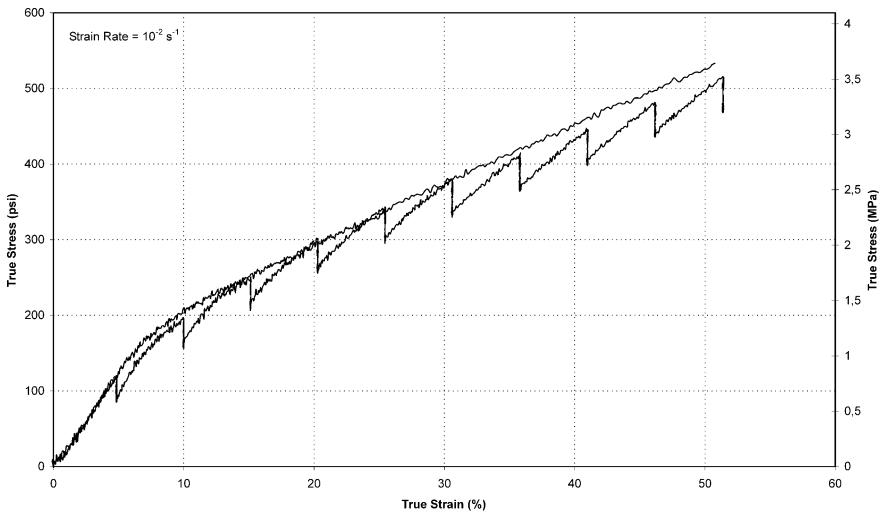


Fig. 14. Multiple step stress relaxation experiments on Adiprene-L100.

desired constant strain rate response. In order to assure this, a multiple step stress relaxation experiment was performed at loading constant strain rate of $\dot{\epsilon} = 10^0 \text{ s}^{-1}$ with relaxation processes at true strain levels of $\epsilon = 15, 30, 45,$ and 60% . Fig. 16 shows the stress–strain relationship from this experiment along with the corresponding constant strain rate behavior. Given that the multiple step stress relaxation behavior followed the constant strain rate response, it is ratified that relaxation processes do not alter the internal structure of the polymer Adiprene-L100.

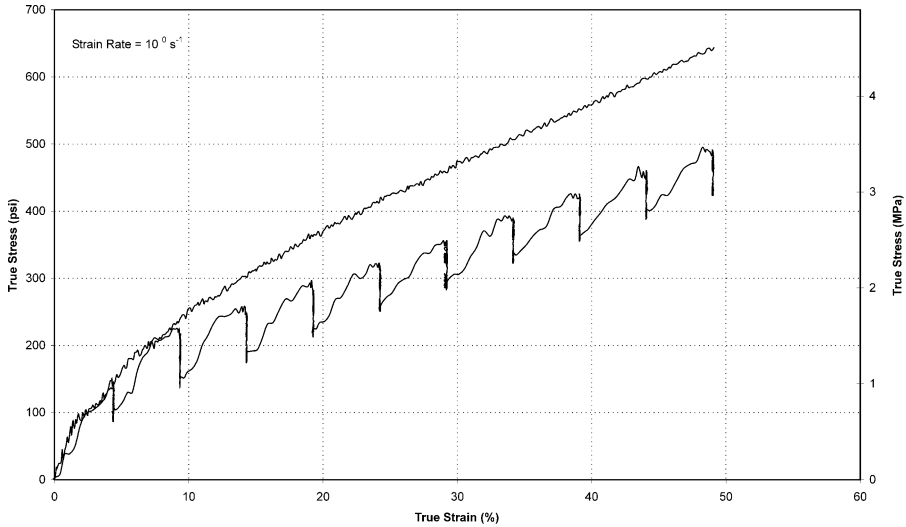


Fig. 15. Multiple step stress relaxation experiments on Adiprene-L100.

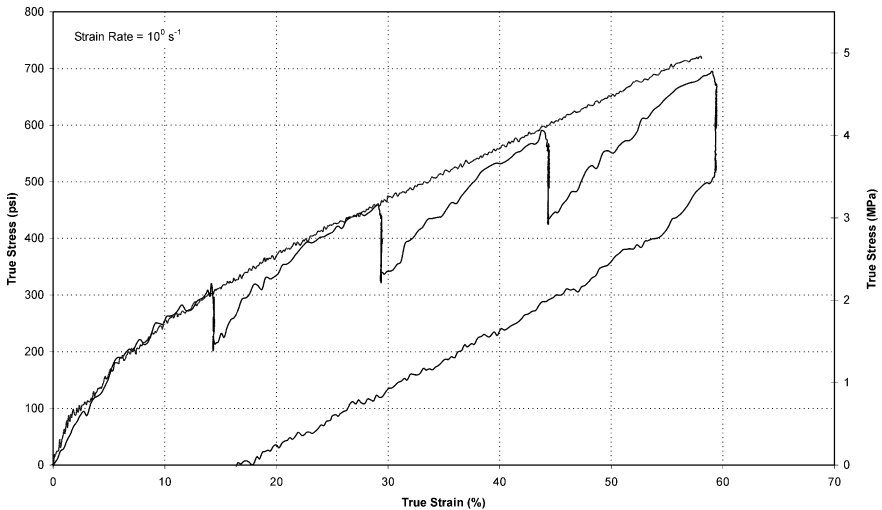


Fig. 16. Multiple step stress relaxation experiments on Adiprene-L100.

4. Conclusions

The comprehensive response of Adiprene-L100 was studied for the first time in the finite deformation regime at different temperatures, different strain rates, different strain levels, and various stress relaxation processes. A strong dependence on temperature was found; the dependence got stronger as the temperature was lowered (colder). As with the majority of polymers, Adiprene-L100 undergoes the glass transition temperature effect. Adiprene-L100 experiences this phenomenon at $T = -80^\circ\text{F}$ (-62°C). A gradual dependence on strain rate was found during quasi-static loading in contradiction to Gray et al. (1997). Similarly to the glass transition temperature effect, Adiprene-L100 exhibited a drastic change from “rubbery” to “glassy” behavior between the strain rates of 5000 and 1 per second. This behavior is not characteristic of all polymers, unlike the glass transition effect. Adiprene undergoes a small but significant plastic deformation during compressive loading. The amount of plastic deformation appeared to depend on strain rate. A decrease in the strain rate resulted in an increase in the permanent deformation. For a strain rate of $\dot{\epsilon} = 10^{-4} \text{ s}^{-1}$ approximately 5% of the total strain was viscoplastic when deformed to a total strain of 55%. As many other polymers, Adiprene-L100 suffers stress relaxation at room temperature. It appears that no changes are introduced by relaxation processes into Adiprene-L100.

References

- Bardenhagen, S.G., Stout, M.G., Gray, G.T., 1997. Three-dimensional, finite deformation, viscoplastic constitutive models for polymeric materials. *Mechanics of Materials* 25, 235–253.
- Khan, A.S., Zhang, H., 2001. Finite deformation of a polymer and constitutive modeling. *International Journal of Plasticity* 17, 1167–1188.
- Kreml, E., 2001. Relaxation behavior and modeling. *International Journal of Plasticity* 17, 1419–2001.
- Gray III, Blumenthal, Carpenter, 1997. Influence of temperature and strain rate on the mechanical behavior of adiprene-1100. Report No: LA-UR-97-0905. Los Alamos National Laboratory.
- McGuirt, C., Lianis, G., 1970. Constitutive equations for viscoelastic solids under finite uniaxial and biaxial deformations. *Trans. Soc. Rheol* 14 (2), 117–134.
- Liang, R., Khan, A.S., 1999. A critical review of experimental results and constitutive models for bcc and FCC metals over a wide range of strain rates and temperatures. *International Journal of Plasticity* 15, 963–980.
- Khan, A.S., Liang, R., 1999. Behaviors of three BCC metal over a wide range of strain rates and temperatures: experiments and modeling. *International Journal of Plasticity* 15, 1089–1109.
- Khan, A.S., Zhang, H., 2000. Mechanically alloyed nanocrystalline iron and copper mixture: behavior and constitutive modeling over a wide range of strain rates. *International Journal of Plasticity* 16, 1477–1492.
- Yuan, Y., Khan, A.S., 2000. Time dependent finite deformation response of a polymer. In: Khan, A.S., Zhang, H., Yuan, Y. (Eds.), *Plastic and Viscoplastic Response of Materials and Metal Forming*. Neat Press, pp. 164–166.
- Lockett, F.J., 1972. *Nonlinear Viscoelastic Solids*. Academic Press, London.