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Viscoelastic–plastic behaviour with mean strain changes in polypropylene

TAKASHI ARIYAMA
Department of Mechanical Engineering, Faculty of Engineering, Science University of Tokyo, 1-3 Kagurazaka, Shinjuku-ku, Tokyo 162, Japan

The stress–strain curves and stress–relaxation curves of polypropylene are obtained by using a closed-loop, electrohydraulic, servo-controlled testing machine. Effects of mean strain changes on deformation behaviour are examined in a tension–compression mode under strain control at room temperature (18–23 °C). The hysteresis loops of three mean strains show a steady-state response from the stress–strain curves at a strain rate of $1 \times 10^{-3}\ \mathrm{s}^{-1}$, at a strain width of 5%, at a number of cycles of $N = 50$ and at three mean strains ($\varepsilon_m = 0, +1.0$ and $+2.0\%$). The drop of stress at the mean strain of $\varepsilon_m = -1.0\%$ is larger in magnitude than that at $\varepsilon_m = +1.0\%$; this is caused by the higher stress level at $\varepsilon_m = -1.0\%$ as compared with the stress level at $\varepsilon_m = +1.0\%$. From the results of stress amplitude and the stress drop behaviour, the magnitude of stress drop is hardly affected by the mean strain.

1. Introduction

The effect of mean strain changes on the cyclic deformation behaviour and morphology of polypropylene has been studied by a closed-looped, electrohydraulic, servo-controlled testing machine [1]. The stress–strain curve data of three mean strains ($\varepsilon_m = 0, +2.5$ and -2.5%) indicated different loop behaviours. At a mean strain of $\varepsilon_m = 0\%$, the peak tensile stress is smaller (ca. 10 MPa) than that of the peak compressive stress and the mean stress was shifted to the compressive side. On the other hand, at $\varepsilon_m = +2.5$ and -2.5%, the mean stress was shifted to the preloading direction. It has been concluded from the results of the stress–strain curves, stress–relaxation curves and SEM fractographs, that microstructural changes occurred significantly under the cyclic preloadings.

Studies on the viscoelastic–plastic properties seem to be important to investigate the feasibility of using the polymeric materials as machinery parts or structural materials. Polypropylene exhibits an excellent fatigue resistance during large strain cyclic deformation [2]. It is reported from the results of the microstructural changes during large strain cyclic deformation of polypropylene, that a neck in tension appeared and that the neck disappeared as the specimen was compressed [3]. The deformation behaviour of ductile polymers, such as polyethylene and polypropylene, is considered to show the different behaviours in tension and compression. The polymers have a cyclic work-softening in cyclic preloadings, including a semi-elastic deformation, and the molecular structure shows complicated features.

On the other hand, the ductile polymer of polycarbonate undergoes a marked decrease in deformation resistance prior to crack formation and bulging of the specimen in the previously necked region. Attala *et al.* [4], in samples of isotactic polypropylene having different morphologies and crystallinities, reported that both crystallinity and spherulite size enhance the non-linearity of the viscoelastic response and that an increase in size of crystalline aggregates increases the values of relaxation moduli.

Cyclic deformation and stress–relaxation behaviour after cyclic loading in isotactic polypropylene was examined in a tension–compression mode [5, 6]. The distinctive shape of the hysteresis loop, termed a propeller-like shape, was characteristic of isotactic polypropylene with large spherulites (180 μm diameter) in marked contrast to the behaviour of some metals [7, 8]. Stress–relaxation behaviours were studied in the polypropylene samples subjected to a cyclic preloading and to a simple tension [9, 10]. The calculated stress–relaxation curves were obtained by the calculation of the constitutive equation based on a linear viscoelastic model, which was a simple three-element model. In the relaxation tests after cyclic preloading, the calculated results agreed very well with the experimental ones. In addition, the calculated stress–relaxation curves were obtained [11, 12] by the calculation of the constitutive equation based on an overstress theory [13], in which the equilibrium stress and the viscosity function were treated. The calculated results agreed very well with the experimental ones. It was concluded that the overstress theory explained the non-linear viscoelastic–plastic properties of polypropylene. On the other hand, Kitagawa *et al.* [14] applied the viscoplasticity theory based on overstress to polypropylene in a servo-controlled torsion machine, where the experimental results agreed well with calculated ones. Bordomaro and Krempl [15] reported the effect of strain rate on the deformation and

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relaxation behaviour of 6/6 nylon at room temperature. They concluded that a unified model with an overstress dependence of the inelastic rate of deformation could be useful for modelling.

The aim of the present study is to analyse the effect of mean strain changes on the cyclic deformation behaviour of polypropylene at room temperature. The stress–strain and stress–relaxation tests were performed under various sets of strain rate, strain amplitude and number of cycles, by use of a closed-loop, electrohydraulic, servo-controlled testing machine.

2. Experimental procedure
2.1. Testing procedure

Stress–strain and stress-relaxation curves were obtained by using a closed-loop, electrohydraulic, servo-controlled testing machine. Applied loads were measured directly by means of a strain gauge load cell. Specimens were tested in a tension–compression mode under strain control at room temperature (18–23 °C). The extensometer was attached to the specimen with two springs lightly acting on two knife edges. Load and displacement mode data were fed to a computer (NEC Co., PC 9801) and were stored on floppy discs after data processing. A detailed description of both the testing machine and the extensometer is given elsewhere [6].

2.2. Materials

The isotactic polypropylene used in this study, Grade J105 in the form of extruded rods with a diameter of 30 mm, was supplied by Ube Industries Ltd. Crystal modification was the monoclinic α-form and a melt flow index of 0.005 kg per 10 min is reported by the supplier. The material has the apparent density of 907 kg m^{-3}, measured by using absolute ethanol at 18 °C. The degree of crystallinity calculated from the density was 56%, where values of 870 kg m^{-3} for completely amorphous density and 936 kg m^{-3} for crystalline density were used [16]. The number average molecular weight was 285 000 g mol^{-1}, which was estimated from the melt flow index. The mean spherulite diameter of the supplied materials was 180 μm [17, 18].

2.3. Sample preparation for cyclic tests

The as-received material was machined to a rod specimen with a lathe. The gauge length was 150 mm and the diameter of the straight part was 8 mm. The surface of the straight part was polished on emery papers because of visible flaws and scratches. The microstructure of the straight part was homogeneous: this fact was confirmed by a direct observation of the spherulite deformation by use of polarizing microscopy [17, 18].

2.4. Conditions of cyclic preloading

Table I lists the conditions of cyclic preloadings. The strain rate ($\dot{\varepsilon}$), number of cycles (N) and mean strain (ε_m) are taken as parameters. The cyclic preloading tests were performed at different strain amplitudes up to $\Delta\varepsilon/2 = \pm 3.5\%$. Three strain rates were chosen ($\dot{\varepsilon}$, s^{-1}): 1×10^{-3}; 1×10^{-4}; 1×10^{-5}. In the strain ranges tested here, no stress whitening was observed with the naked eye.

TABLE I Conditions of cyclic preloadings

Number of cycles N	50
Strain rate $\dot{\varepsilon}$ (s^{-1})	10^{-3}, 10^{-4}
Mean strain ε_m (%)	0, + 1.0, + 2.0, + 4.0
Strain width $\Delta\varepsilon$ (%)	3.0, 5.0, 7.0

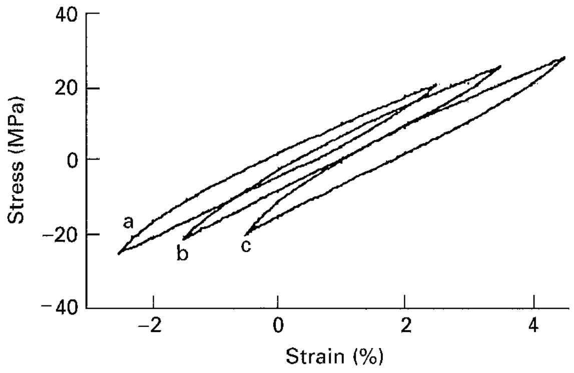

Figure 1 Stress–strain curves at a strain rate of 1×10^{-3} s^{-1}, at a strain width of 5%, at a number of cycles of $N = 50$ and at three mean strains (ε_m, %): a, 0; b, + 1.0; c, + 2.0.

2.5. Stress–relaxation tests

The stress–relaxation tests were performed after three cycles (N): 35; 45; 50; at three mean strains (ε_m): + 1; + 2; + 4%. The tests were made after different predetermined strain rates, at a testing time of $t = 1200$ s for all tests.

3. Results and discussion
3.1. Stress–strain behaviour

Fig. 1 shows the stress-strain curves at a strain rate of 1×10^{-3} s^{-1}, at a strain width of 5%, at a number of cycles of $N = 50$ and at three mean strains of $\varepsilon_m = 0$, + 1.0 and + 2.0%. The hysteresis loops of three mean strains show a steady-state response. Cyclic deformation in polypropylene has been studied on the basis of experimental stress–strain curves under various sets of strain rate, number of cycles and strain amplitude [5, 6]. These stress–strain curves were obtained at a mean strain of $\varepsilon_m = 0\%$. The distinctive shape of the hysteresis loop, which was propeller-like, was characteristic of polypropylene with comparatively large spherulites, in contrast to the loops observed in other polymers such as high-impact polystyrene and the acrylonitrile–butadiene–styrene copolymer (ABS) [19], and in some metals like austenitic stainless (SUS 304) and ferritic 2.25Cr-1Mo

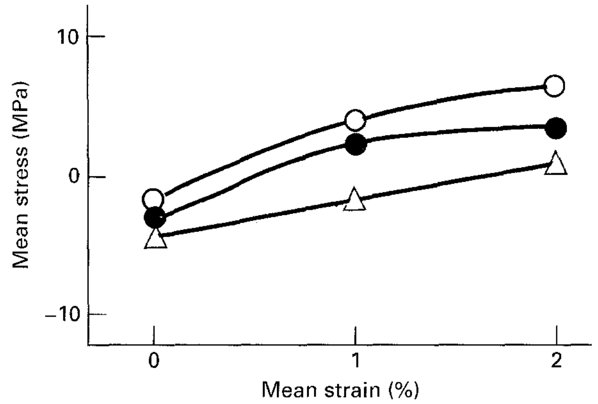

Figure 2 Mean stress versus mean strain at different strain widths (%): ○, 3; ●, 5; △, 7.

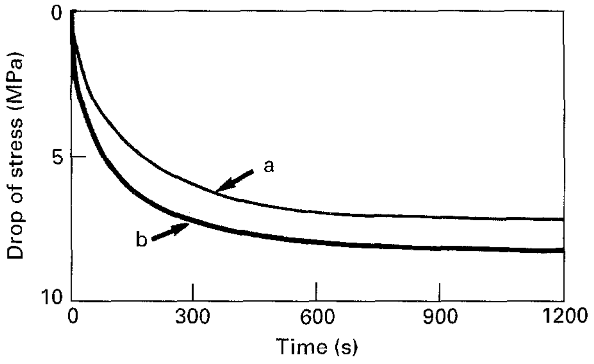

Figure 3 Stress–relaxation curves at a strain rate of 1×10^{-3} s^{-1}, at a strain width of 7%, after a number of cycles of $N = 50$ and at different mean strains (ε_m, %): a, + 1.0; b, − 1.0.

steels [8]. The propeller-like loops changed shape into regular loops (steady-state loops) as the number of cycles was increased up to $N = 30$–50. The change in loop shape was found to be due to the interaction between the molecular chains within the spherulites and the chains of the amorphous parts. It was seen that at strain amplitudes $> 1.5\%$, the magnitude of stress amplitude (or cyclic stress width) decreased with increasing the number of cycles up to $N = 30$; while at strain amplitudes $< 1.5\%$, the magnitude of stress amplitude was nearly constant. Thus, it appeared that at higher strain amplitudes, a stable molecular arrangement was formed because of release of previously existing strain.

Both the peak tensile stress and peak compressive stress increase with increasing mean strain, as shown in Fig. 1. The hysteresis loop shifts to the upper side of the stress axis. Fig. 2 shows the mean stress versus mean strain at three strain widths of 3, 5 and 7%. The mean stress is defined as the mean of the peak tensile stress and peak compressive stress. The mean stress increases with increasing mean strain. The stress increment between $\varepsilon_m = 0$ and 1.0% is larger than that between $\varepsilon_m = 1.0$ and 2.0%. This fact therefore shows a strong effect of mean strain on the stress–strain behaviour as the mean strain approaches zero. The work hardening coefficient (E_p) on the tensile side is slightly smaller than that on the compressive side at three strain widths of 3, 5 and 7%. E_p is defined as the ratio of stress : strain in the plastic region. The as-received polypropylene sample is essentially isotropic, as manifested by wide-angle X-ray diffraction and polarizing microscopy [20], and is composed of spherulites. The lamella in the spherulites radiate outwards from the centre of the spherulite. The tension is expected to exert the scission of molecular chains or the scission of amorphous parts in the spherulites. The viscosity on the tensile side is lowered by cyclic formation. In contrast, little microstructural change is observed in the case of the compression of cyclic loadings.

3.2. Stress-relaxation behaviour

Fig. 3 shows the stress–relaxation curves at a strain rate of $1 \times 10^{-3}\,s^{-1}$, at a strain width of 7%, after a number of cycles of $N = 50$ and at different mean strains of $\varepsilon_m = +1.0$ and -1.0%. The drop of stress at the mean strain of $\varepsilon_m = -1.0\%$ is larger in magnitude than that at $\varepsilon_m = +1.0\%$. This is caused by the higher stress level at $\varepsilon_m = -1.0\%$ as compared with the stress level at $\varepsilon_m = +1.0\%$. In addition, both stress–relaxation curves reach an equilibrium stress (or steady state) at about duration time $t = 800$ s.

The stress drop as a function of testing time after the simple (monotonical) tension ($N = 0$) was obtained at different strains of $\varepsilon = 1$, 3 and 5% [10]. The experimental data revealed that the stress drop behaviour at $N = 0$ was very sensitive to strain ε. In addition, the stress drop at $N = 0$ increased in the order of increasing strain. At the lowest strain of $\varepsilon = 1\%$, the stress drop behaviour showed similar behaviour. The magnitude of the stress drop at strains of $\varepsilon = 3$ and 5% was larger than that at $\varepsilon = 1\%$. Moreover, the stress drop as a function of testing time in the cyclically preloaded samples was obtained at different strain amplitudes [10]. The stress drop behaviour after cyclic preloading ($N = 50$) was significantly different from that after simple tension ($N = 0$), i.e. the amount of stress drop was smaller when the samples were subjected to cyclic preloading [10]. Moreover, the stress drop increased with an increase in strain amplitude in the same fashion as with the simple tension. These findings are explained in terms of the formation of a stable molecular rearrangement: in fact, the hysteresis loop after the cyclic preloading of $N = 50$ tended to reach a steady state [5].

Figs 4 and 5 show the stress–relaxation curves at a strain rate of $1 \times 10^{-3}\,s^{-1}$, at strain widths of 3, 5 and 7%, after a number of cycles of $N = 50$, at mean strains of $\varepsilon_m = +1.0$ and $+2.0\%$, respectively. In the cases of $\varepsilon_m = +1.0$ and $+2.0\%$, the magnitude of the stress drop increases as the strain width increases. A small difference between the stress drop at $\varepsilon_m = +1.0$ and $+2.0\%$ is observed at strain width of 5%. At strain widths of 3 and 5%, however, the magnitude of the stress drop is almost the same.

Figs 6 and 7 show the stress–strain curves at a number of cycles of $N = 50$ and at mean strains of $\varepsilon_m = +1.0$ and $+2.0\%$, respectively. Large differences in the stress amplitude is not seen at each mean strain. From this result, and the aforementioned stress

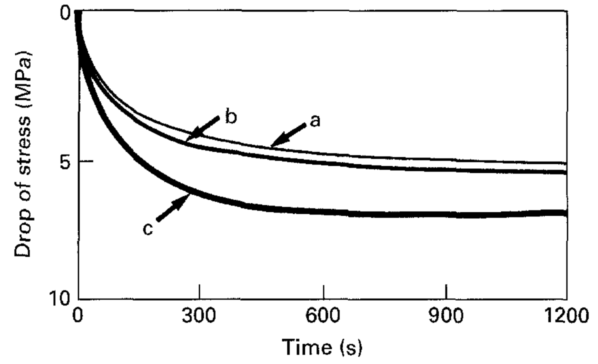

Figure 4 Stress–rexalation curves at a strain rate of 1×10^{-3} s^{-1}, at a mean strain of $\varepsilon_m = +1.0\%$, after a number of cycles of $N = 50$ and at different strain widths (%): a, 3; b, 5; c, 7.

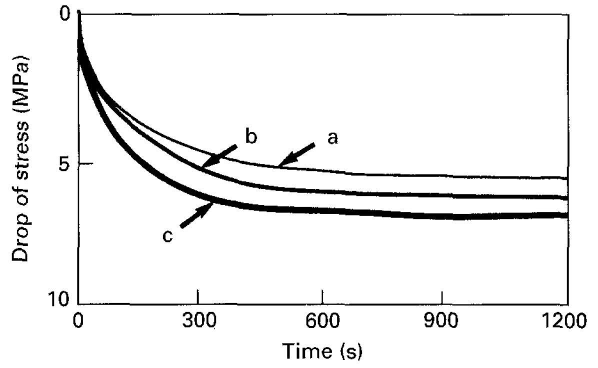

Figure 5 Stress–relaxation curves at a strain rate of 1×10^{-3} s^{-1}, at a mean strain of $\varepsilon_m = +2.0\%$, after a number of cycles of $N = 50$ and at different strain widths (%): a, 3; b, 5; c, 7.

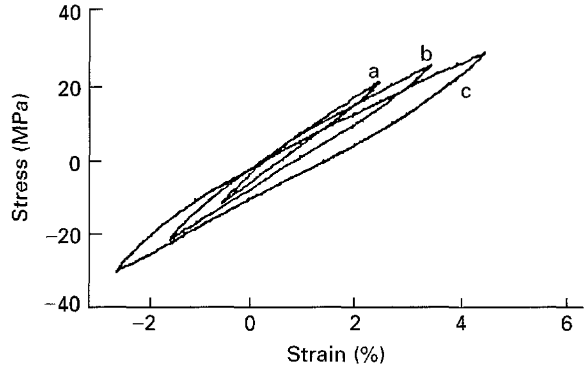

Figure 6 Stress–strain curves at a number of cycles of $N = 50$, at a mean strain of $\varepsilon_m = +1.0\%$ and at different strain widths (%): a, 3; b, 5; c, 7.

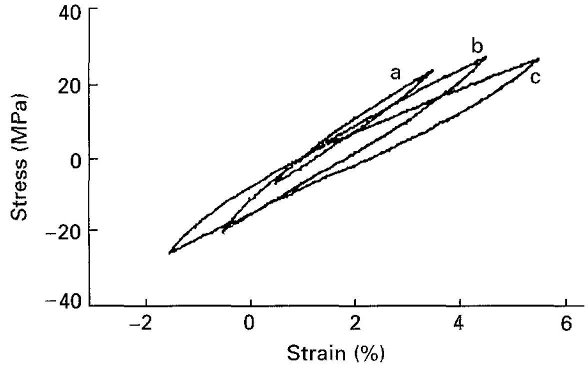

Figure 7 Stress–strain curves at a number of cycles of $N = 50$, at a mean strain of $\varepsilon_m = +2.0\%$ and at different strain widths (%): a, 3; b, 5; c, 7.

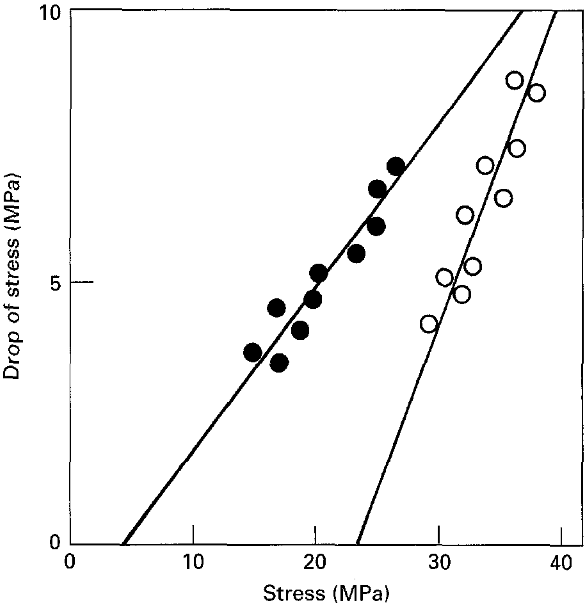

Figure 8 Drop of stress at the testing time of $t = 1200$ s versus stress level at the beginning of stress– relaxation at different mean strains (ε_m, %): ●, + 1.0; ○, − 1.0.

drop behaviour, the magnitude of stress drop is hardly affected by the mean strain.

Fig. 8 shows the drop of stress at the testing time of $t = 1200$ s versus stress level at the beginning of stress–relaxation at mean strains of $\varepsilon_m = +1.0$ and -1.0%. Two solid lines represent the calculated values determined from the experimental data by using a method of least squares. The drop stress on both the tensile ($\varepsilon_m = +1.0\%$) and compressive ($\varepsilon_m = -1.0\%$) sides increases in a linear fashion with increasing stress level. The gradient on the compressive side is greater than that on the tensile side. Accordingly, the mean strain of $\varepsilon_m = -1.0\%$ appears to exert influence to reduce viscoelastic characteristics. It is reported [10] that the relationship between the stress drop and the stress level after simple tension ($N = 0$) and after cyclic preloading ($N = 50$) showed good linearity with different slopes. The gradient at a strain rate of 1×10^{-4} s^{-1} and a number of cycles of $N = 0$ had the greatest value; on the contrary, the gradient at a strain rate of 1×10^{-3} s^{-1} and $N = 50$ had the least value. In addition, the amount of stress drop increased with increasing strain amplitude and with increasing stress level. It has been concluded that cyclic preloading exerted on effect on a reduction in viscoelastic properties.

4. Conclusions

Effects of mean strain changes on deformation behaviour of polypropylene are examined in a tension–compression mode under strain control at room temperature (18–23 °C) by using a closed-loop, electrohydraulic, servo-controlled testing machine. From the stress–strain curves at a strain rate of 1×10^{-3} s^{-1}, a strain width of 5%, at a number of cycles of $N = 50$ and at three mean strains of $\varepsilon_m = 0$, $+1.0$ and $+2.0\%$, the hysteresis loops of three mean strains show a steady-state response. The drop of stress at the mean strain of $\varepsilon_m = -1.0\%$ is larger in magnitude than that at $\varepsilon_m = +1.0\%$; this is caused by the higher stress level at $\varepsilon_m = -1.0\%$ as compared with the stress level at $\varepsilon_m = +1.0\%$. Large differences in the stress amplitude are not seen at each mean strain. From this result, and the stress drop behaviour, the magnitude of stress drop is hardly affected by the mean strain. Cyclic preloading exerts on effect on a reduction in viscoelastic properties. From the view point of the stress level, the strain width and/or the strain rate affect the deformation characteristics of polypropylene as compared with mean strain.

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