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Relationships between nonlinear dynamic viscoelasticity and fatigue behaviors of glassy polymer under various fatigue test conditions

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Summary

Fatigue behavior of amorphous polystyrene (PS) under various cyclic strain conditions was investigated on the basis of nonlinear dynamic viscoelastic measurement at various ambient temperatures. The degree of nonlinear viscoelasticity under tension-compression type cyclic strain was greater than that under tension or compression type ones. The longest fatigue lifetime and the shortest one were observed under compression and tension type fatigue tests, respectively. The fatigue lifetime decreased with an increase in nonlinear viscoelastic parameter (NVP) in the case of the same type of cyclic strain conditions.

Introduction

Though polymeric materials show great differences in mechanical properties from metallic materials because of their viscoelastic natures, most studies on the fatigue behavior of polymeric materials still follow the methods that have been developed for metals. The authors made an attempt to reveal the relationship between nonlinear dynamic viscoelastic characteristics and fatigue behavior in the cases of crystalline polymers [1-3]. However, as to glassy polymers, such relationship still remains unknown. Also, the effect of cyclic strain conditions on nonlinear dynamic viscoelastic characteristics and fatigue behavior for polymeric materials has not been extensively clarified yet.

In this study, the nonlinear dynamic viscoelastic characteristics of amorphous polystyrene were measured under various fatigue test conditions, and the relationships among the types of cyclic strain conditions, nonlinear dynamic viscoelastic characteristics and fatigue behavior were discussed.

Experimental

Materials

Polystyrene (PS) (M_w=338,000, Idemitsu Petrochemical Co., Ltd.) was used as the specimens of fatigue test. The specimens for fatigue tests were compression-molded into a

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dumbbell-shaped mold with 5 mm in width, 3 mm in thickness and 20 mm in gauge length.

Nonlinear dynamic viscoelastic measurement and fatigue test

Fig.1 shows the blockdiagram of the fatigue tester to investigate nonlinear dynamic viscoelastic behavior under various fatigue test conditions [1, 3]. The imposed dynamic strain, $\varepsilon(t)$ was a sinusoidal one with the amplitude of ε_d and the angular frequency of ω as expressed by eq (1). Its stress response signal, $\sigma(t)$ was converted into digital data with a high-speed A/D converter and then, these digital data were transformed into Fourier series as expressed by eq (2). The nonlinear viscoelastic parameter, NVP was calculated by the coefficients of the Fourier series of stress response as given in eq (3) [1-3].

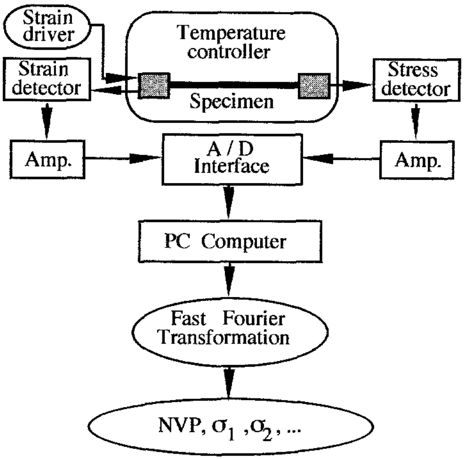


Fig. 1 Blockdiagram of fatigue tester for investigation of nonlinear dynamic viscoelastic properties of polymeric materials under cyclic fatigue

$$\varepsilon(t) = \varepsilon_d \sin(\omega t) \tag{1}$$

$$\sigma(t) = \sigma_1 \sin(\omega t + \delta_1) + \sigma_2 \sin(2\omega t + \delta_2) + \ldots + \sigma_n \sin(n\omega t + \delta_n) \tag{2}$$

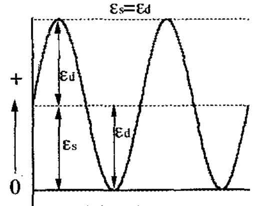
$$\mathrm{NVP} = \frac{\sigma_2 + \sigma_3 + \ldots \ldots \sigma_n}{\sigma_1} \tag{3}$$

where σ_1 and δ_1 are the fundamental stress amplitude and phase angle, respectively. $\sigma_2, \sigma_3, \ldots, \sigma_n$ and $\delta_2, \delta_3, \ldots, \delta_n$ are the higher harmonic stress amplitudes and phase angles, respectively. The number of higher harmonics, n was taken up to 100 since the higher harmonics over 100th were very small and could be ignored. In order to investigate the effect of cyclic strain conditions on fatigue behavior for glassy polystyrene, three types of cyclic strain conditions, tension, compression and tension-compression types were employed in this study as shown in Fig.2. In the cases of the tension, the tension-compression and the compression type cyclic strain conditions, the static strain, ε_s are equal to the dynamic strain amplitude $\varepsilon_d (\varepsilon_s = \varepsilon_d)$, zero $(\varepsilon_s = 0)$, and the negative value of dynamic strain amplitude ε_d $(-\varepsilon_s = \varepsilon_d)$, respectively. The fatigue tests were carried out at the ambient temperatures of 303K, 333K and 363K with frequency of 9.26Hz.

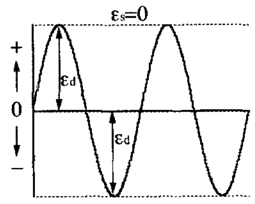
Results and discussion

Relationship between nonlinear dynamic viscoelasticity and types of cyclic strain

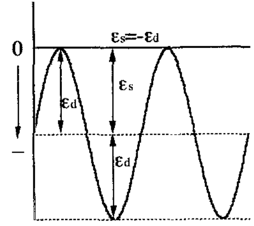
Fig.3 shows the relationships between magnitude of NVP and types of cyclic strain for PS under two different dynamic strain amplitudes at 303K. The greatest nonlinear dynamic viscoelastic behavior was observed under the tension-compression cyclic strain. The mechanical properties of polymers under the tensile deformation condition are generally different from those under the compressive deformation condition [4]. Fig.4 and Tab.1 show the stress-strain curve and the mechanical properties of PS under the uniaxial tensile and compressive deformation, respectively. Because the compressive modulus of PS was larger than the tensile one as shown in Tab.1, the stress response in the compressive region during one deformation cycle must be larger than that in the tensile region in the case of tension-compression type cyclic strain. Then, the tension-compression cyclic strain would induce the unsymmetrical stress response with a larger compressive response and smaller tensile one. Therefore, it seems reasonable to consider that the strong nonlinear dynamic viscoelasticity might arise from the remarkably unsymmetrical wave of stress response in the case of tension-compression type cyclic strain.



(a) Tension type



(b) Tension-compression type



(c) Compression type

+: tensile direction

-: compressive direction

Fig.2 Three types of cyclic strain

Also, since the strain dependence of stress for PS became more sensitive in the compressive region of larger strain as shown in Fig.4, the magnitude of NVP under the compression type cyclic strain might increase more steeply with an increase in the dynamic strain amplitude as shown in Fig.3. In the case of the fatigue test under the tension type cyclic strain, the magnitude of NVP did not show a distinct increase with an increase in strain amplitude, maybe due to a near linear stress-strain relationship in a wider tension region as shown in Fig.4. Therefore, Figs. 3 and 4 apparently indicate that the nonlinearity of stress-strain curve gives a remarkable effect on the nonlinear dynamic viscoelasticity for glassy polymers such as PS.

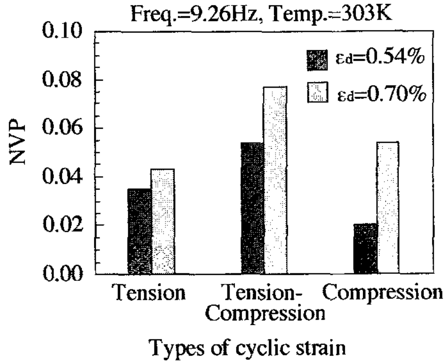


Fig.3 Relationships between NVP and types of cyclic strain under various dynamic strain amplitudes after 100 seconds from the start of fatigue test at 303K

Fig.5 shows the relationships between NVP and the types of cyclic strain at three different temperatures for PS under the condition of the dynamic strain amplitude of 0.54%. The magnitude of NVP increased with temperature in the temperature range studied here, apparently in the cases of tension and tension-compression type cyclic strain, but slightly in the case of compression type cyclic strain. Since the temperature is close to the glass transition temperature of 363K, the temperature dependence of modulus for the tension and the tension-compression cyclic strain is more remarkable than that for the compression one. Then, the irreversible changes in aggregation structure might be accumulated more remarkably during tension and tension-compression type cyclic strain than compression one. The irreversible structural changes might cause that the magnitude of NVP became larger and increased apparently with temperature under the tension and the tension-compression type cyclic strain.

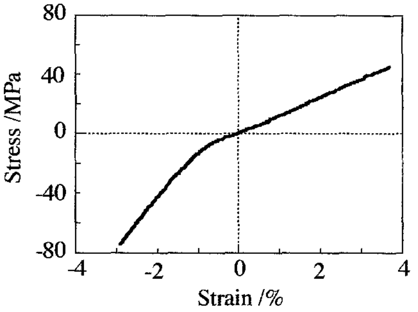


Fig. 4 Stress-strain curve of PS under tension and compression conditions

Tab.1 Mechanical properties of PS under tension and compresion conditions

	E /GPa	σ_b /MPa	ε_b /%
Tension	1.2	45	3.7
Compression	2.5	-78	-2.9

E: tensile modulus; σ_b: break strength; ε_b: break strain

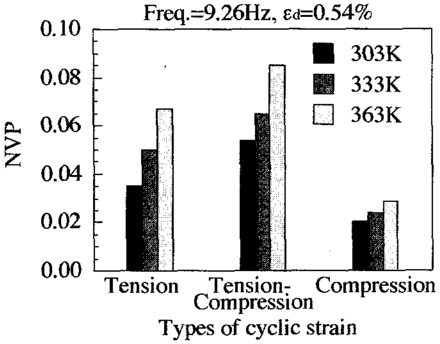
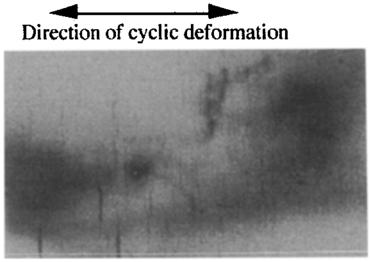
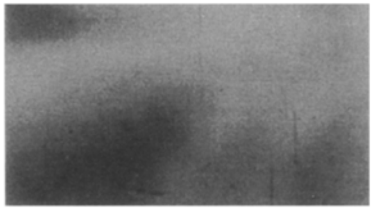


Fig.5 Relationships between NVP and types of cyclic strain under various temperatures after 100 seconds from the start of fatigue test

The craze formation during fatigue test for glassy polymers such as PS has been investigated by many groups [4-8]. The visible crazes or macrocrazes were considered to be initiated from the microcrazes with invisible dimensions. The concentration of microcrazes initiated during fatigue test was evaluated as high as 10^{15} cm^{-3}, uniformly distributed throughout the specimen [6]. However, only a few of these invisible microcrazes were able to grow up to a visible size which could be observed by microscope or even by naked eyes. In the case of glassy polymers, the dimension of crazes was generally 0.1-1.0 μm in width, 0.1-0.5 μm in depth, and the length could extend as long as 10 mm [7]. Fig.6 is the optical micrographs of the crazes formed on PS films after being subjected to a fatigue test for 1,000 seconds at 363K. No visible crazes were observed on PS films after being subjected to the fatigue test for 1,000 seconds at 303K and 333K. Also, though visible crazes were not observed under the compression type cyclic strain at 363K, the apparent crazes on PS film were observed under the tension and the tension-compression type cyclic strain at 363K as shown in Fig.6. It has been found that the irreversible changes in aggregation structures for polymeric materials occurred during the fatigue test with larger imposed strain amplitude [3]. However, these irreversible structural changes did not always develop into visible crazes in all fatigue test conditions. In the cases of the tension and the tension-compression type of cyclic strain, the formation of visible crazes becomes more possible since the cyclic tensile force accumulates the growth of microcrazes. This led to the apparent visible crazes which were observed under tension and tension-compression types of cyclic strain. However, in the case of compression type cyclic strain, since the cyclic compressive force could not accumulate the growth of microcrazes as the tensile one, no apparent visible crazes were observed under compression type cyclic strain. Also, in oder to form visible crazes, it is necessary that either the disentanglement of molecular chains or the chain fracture occur in the region of crazes [4, 5]. Since the disentanglement of molecular chains became more remarkable at the temperature close to glass transition temperature due to the activation of thermal molecular motion of polymeric chains in this temperature region, apparent crazes on PS films were observed at 363K rather than at 303K or 333K under the tension and the tension-compression type cyclic strain with the imposed strain amplitude of 0.54%.



(a) Tension type



(b) Tension-Compression type

Fig.6 Optical micrographs of PS after 1000 seconds from the start of fatigue tests under tension type (a) and tension-compression type (b) cyclic strain conditions at 363K under imposed strain amplitude of 0.54%

On the other hand, in the cases of tension and tension-compression type cyclic strain, the magnitudes of NVP were much larger than that in the compression one, specially at higher temperature region (Fig.5). As discussed in our precious paper [3], if a specimen exhibits a larger NVP, the irreversible structural changes will occur more frequently during cyclic fatigue test. So the large magnitude of NVP under tension and tension-compression type fatigue tests indicated that the irreversible structural changes would occur more frequently under tension and tension-compression type fatigue tests than that under compression one. Furthermore, it may be reasonable to consider that the formation of crazes for glassy polymers is related to the remarkable nonlinear dynamic viscoelastic behavior; i. e. the large NVP.

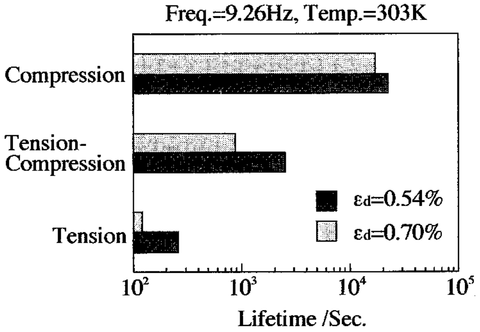


Fig.7 Relationship between the types of cyclic strain and fatigue lifetime under imposed strain amplitudes of 0.54% and 0.70% at 303K

Fatigue behavior under different types of cyclic strain

Fig.7 shows the relationships between the types of fatigue tests and fatigue lifetime for PS under the imposed strain amplitudes of 0.54% and 0.70% at 303K. The shortest fatigue lifetime and the longest one were observed under the tension type and the compression type fatigue tests, respectively.

The shortest fatigue lifetime under the tension type fatigue test could be attributed to that the tensile force was favorable to the formation or the growth of crazes, and the longest fatigue lifetime under the compression type one could be attributed to that the compressive force was unfavorable to the formation or the growth of crazes.

On the other hand, according to the results from Figs.3 and 7, it could be found that the fatigue lifetime decreased with an increase in the magnitude of NVP under the same type fatigue test. This result was well consistent with the conclusion of our previous reports [1-3], that is, the fatigue strength of polymeric materials decreased with an increase in the magnitude of NVP in the case of tension fatigue test. Hereupon, it should be noticed that the fatigue lifetime under the tension type cyclic strain was shorter than that under the tension-compression one though the latter showed greater magnitude of NVP at the same strain amplitude. Therefore, it is able to conclude that such fatigue lifetime - NVP relationship can be applied only under the same type of fatigue test since the dynamic stress-strain behavior of polymeric solids should be strongly dependent on the type of cyclic strain.

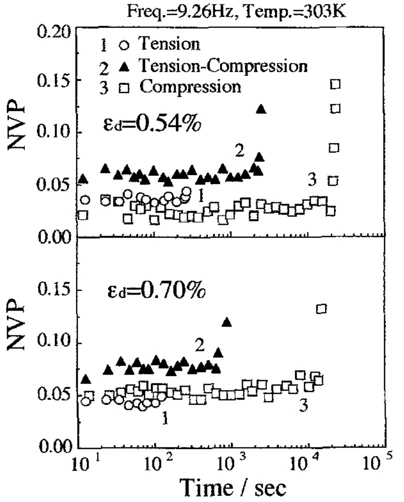


Fig.8 Variations of NVP with time during fatigue test under various cyclic strain conditions and various dynamic strain amplitudes

Fig.8 shows the variations of NVP with time during fatigue test for PS under different types of fatigue test with various dynamic strain amplitudes at 303K. The nonlinear dynamic viscoelastic behavior showed no significant changes until very close to fatigue failure under each type of fatigue test. When the fatigue test approached to the point of fatigue failure, an apparent increase in the magnitude of NVP was observed, especially under the compression type fatigue test. This might arise from the local irreversible structural changes corresponding to the onset of fatigue failure [1-3].

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