

Creep Rupture Properties of Homopolymer, Copolymer, and Terpolymer Based on Poly(oxymethylene)

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ABSTRACT: Polyacetal copolymers were prepared by cationic ring-opening copolymerizations of 1,3,5-trioxane (TOX) with 1,3-dioxolane (DOX), and polyacetal terpolymers were prepared by terpolymerizations of TOX, DOX, and 2-ethylhexyl glycidyl ether (EHGE). Polyacetal polymers with three different structures such as polyacetal homopolymers, polyacetal copolymers, and polyacetal terpolymers were compared in the mechanical properties and the creep characteristics, and discussed from the view point of the polymer structure. The polyacetal copolymers and the polyacetal terpolymers were determined by ¹H-MNR measurement. About 80 mol % of DOX and EHGE amounts in feed were incorporated randomly into the each polymer. From the plots of the degree of crystallinity (X_c) versus the tensile strength, the tensile strength and crystallinity of the polyacetal homopolymers are higher than those of the polyacetal copolymers and the polyacetal terpolymers. However, the tensile strength does not

decrease linearly with a decrease in the crystallinity among the polyacetal polymers with three different structures, the polyacetal homopolymer, the polyacetal copolymers, and the polyacetal terpolymers. Creep rupture was characterized by the activation volume, v_c , value in Zhurkov's equation, which can be estimated from the slope in the plots of load versus log (rupture time) at 80°C. The polyacetal polymers with higher molecular weight have larger values of the activation volume than those with lower molecular weight. When the activation volume values are compared among the polyacetal polymers with the same molecular weights, they increase in the following order: the polyacetal homopolymers < the polyacetal copolymers < polyacetal terpolymers. © 2010 Wiley Periodicals, Inc. *J Appl Polym Sci* 116: 3242–3248, 2010

Key words: thermoplastics; thermodynamics; creep; structure; fracture

INTRODUCTION

Polyacetal homopolymer (polyoxymethylene, POM) is well-known as a pioneer of engineering plastics since manufacture and sale by DuPont as Delrin (polyacetal homopolymer) in 1960 and followed by Celanese Co. as Celcon (polyacetal copolymer) in 1961. Polyacetal polymers have excellent properties such as good friction wear property, good mechanical and physical properties, chemical resistance and the creep durability. For these excellent properties of the polyacetal polymers, they have been applied to mechanical component parts such as cogwheels and gears, vehicles, an electronic item, and various machines.¹ In general, polyacetal homopolymers could be obtained by anionic polymerization of formaldehyde, followed by end-capping reaction (esterification) with acetic anhydride and so on.² This method has been used to suppress easy depoly-

merization of the polyacetal homopolymer. On the other hand, the polyacetal copolymers are obtained by cationic copolymerization of trioxane with comonomer such as cyclic ethers and cyclic formals, and then are subjected to thermal treatment after copolymerization to remove the unstable ends of the polymers.³ The advantage in the production of polyacetal copolymer is possible to introduce the stable end into the polymer backbone. The polyacetal copolymers have excellent properties in molding, the heat stability, and a long-term mechanical and physical properties, though they have lower crystallinity and lower short-term mechanical strength in comparison with polyacetal homopolymers.⁴ Generally, a long-term mechanical and physical property, especially the creep property, for polyacetal polymers is an important factor to keep the reliability for the usage as mechanical components and mechanical parts in vehicles.

The response of a polymeric material to mechanical deformation and rupture behavior reflects the microscopic deformation processes occurring on a molecule or atomic level, but the phenomenon is complex.⁵ The creep rupture as one of long-term physical properties of the polyacetal polymers is

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presumed that the ductile fracture by the shear yielding is predominant.⁶ Especially the rupture behavior of a crystalline polymer is strongly influenced by the higher-order crystal structure or the molecular cohesion, which are dependent upon the temperature and the deformation speed.^{7,8}

There are already some research papers about the creep rupture of the polyacetal polymers (homopolymers and copolymers).^{9,10} However, the comparison between the polyacetal homopolymers and the polyacetal copolymers remains unclear in detail. Especially the effect of the kinds of the co-monomer on the creep rupture has not been studied.

In this work, the mechanical properties and the creep characteristics of polyacetal polymers are compared in three different structures, polyacetal homopolymers, polyacetal copolymers, and polyacetal terpolymers and discussed on the basis of the polymer structure.

EXPERIMENTAL

Preparation of polyacetal polymers

Commercial poly(oxymethylene) with two different kinds of molecular weights (Delrin 100P and 500P, produced by DuPont Co.) were used as samples. Polyacetal copolymers were prepared by the cationic copolymerization of 1,3,5-trioxane (TOX) with 1,3-dioxolane (DOX), both of which were supplied by Polypastics (Shizuoka, Japan), using boron trifluoride (BF₃) as a catalyst. A continuous-type reactor was used for the polymerization and the reactor temperature was controlled at 80°C. In the polymerization, a small amount of dimethoxymethane (DMM) as a chain-transfer agent was used to adjust the molecular weight and shear viscosity of the polyacetal copolymer and the polyacetal terpolymer. Raw polymers in the reactor were poured into a large amount of an aqueous solution containing a triethyl amine and then the precipitated polymers were left for 12 h at room temperature to deactivate the catalyst. The resulting polymers were post-treated by using an extruder at 200°C to remove unstable hemiformal fractions present in the polymers, and then were formed into pellets. Polyacetal terpolymers were prepared by the cationic terpolymerization of TOX, DOX, and 2-ethylhexyl glycidyl ether (EHGE: Wako Pure Chemical Industries) and formed into pellets by the same post-treatment mentioned above.

Characterization

¹H-NMR spectra of the polyacetal copolymers and the polyacetal terpolymers were taken on a Bruker Avance 400 spectrometer (Switzerland) using

1,1,1,3,3,3-hexafluoro-2-propanol-*d*₂ (HFIP-*d*₂) with trimethylsilane as an internal standard at 35°C. The samples for the NMR measurement were purified by dissolution-precipitation method by using HFIP as a solvent and methanol as a precipitant, respectively. The DOX content in the polyacetal copolymers and the DOX and EHGE contents in the polyacetal terpolymers were determined by ¹H-NMR spectrometry according to the same methods reported previously.^{11–13} The signals of the —CH₂CH₂O— group assigned to the DOX unit and the —CH₂CH₂CH₃ group assigned to the EHGE unit were detected in the spectrum of ¹H-NMR, and the each unit content was calculated from the area of each characteristic peak.

Preparation of injection-molded specimens

The test pieces are molded in a ASTM-D638 dumb-bell type [50 mm (length) × 12 mm (width) × 3 mm (thickness)] by using injection molding machine on Toshiba IS80. On molding, cylinders and nozzle are kept at the temperature of 200°C, the metallic shell's temperature is kept at 80°C to obtain specimens by injection, and the shooting speed is 20 cm³/s, the shooting pressures is 50 MPa and the cooling time is 20 sec.

Evaluation of creep and tensile properties

The creep test was carried out by using the above-mentioned tensile test pieces at 80°C on A&D Co. creep test machine. The tensile strength and elongation at break were measured according to ISO 527–1,2. Tensile tests were performed on an Orientech RTM100 testing machine at 23°C and 50% relative humidity and the specimens were kept under the same conditions for more than 48 h before the test.

Molecular weight measurement

The weight-average molecular weight (M_w) and polydispersity index (M_w/M_n) of the commercial polyacetals were determined by size exclusion chromatography (SEC) on TOSOH HLC-8120GPC using HFIP as an eluent at a flow rate 0.5 mL/min at 40°C and poly(methyl methacrylate) (PMMA) standards for calibration. And also, the melt index (MI), which was measured at 190°C on a Technol Seven Co. Mel-tindexer L-202, was used as the viscosity under a constant stress.

Crystal structure analysis

The polyacetal samples were prepared by cutting the center part of the test piece in 1 mm thickness film. The degree of crystallinity of the polyacetal polymers was calculated on the basis of the diffraction pattern intensity obtained by the wide-angle X-

TABLE I
Polymerizations of the Polyacetal Copolymers and the Polyacetal Terpolymers

Sample	Monomer in feed					Yield (%)	Polymer composition ^a		Molecular structure ^b		
	TOX (mol %)	DOX (mol %)	EHGE (mol %)	BF ₃ (ppm)	DMM (ppm)		DOX (mol %)	EHGE (mol %)	$M_w/10^4$	M_w/M_n	MI/(g/10 min)
Copolymer											
Cop-1	95.8	4.2	–	25	500	55	4.0	–	9.78	1.9	2.5
Cop-2	95.8	4.2	–	25	660	52	4.0	–	6.82	1.8	9.0
Cop-3	95.0	5.0	–	32	600	51	4.7	–	6.90	1.9	8.9
Terpolymer											
Ter-1	95.8	4.1	0.13	38	430	50	3.9	0.1	9.52	2.0	2.8
Ter-2	95.8	4.0	0.15	42	580	48	3.9	0.1	6.77	1.9	9.2
Homopolymer											
100P									10.4	2.0	2.0
500P									6.02	1.9	15.0

^a Determined by ¹H-NMR measurement.

^b Determined by SEC measurement.

ray diffraction (WAXD) measurement, performed on Rigaku Rint1400 with CuK α radiation at an accelerating voltage of 50 kV and a current of 120 mA. Volume fraction of the crystalline region (X_c) was calculated from the density and from the following equation (1):

$$X_c = (\rho - \rho_a) / (\rho_c - \rho_a) \quad (1)$$

where $\rho_c = 1.49$ g/cc is a density of the crystalline region, $\rho_a = 1.28$ g/cc is a density of the amorphous region, and ρ is a density of the samples for the polyacetals.^{14–16}

The lamella periodicity (l_e) was obtained on the basis of the Bragg interval of the most lowest scattered peak in the 2 θ -I profile by small-angle X-ray scattering (SAXS) measurement, performed on RINT 2500HF, and it was calculated from equation (2):

$$l_e = 2\pi/q = \lambda/2 \sin \theta \quad (2)$$

where λ is a wavelength of the incident X-ray beam of 0.15418 nm, q is a scattering vector defined as $q = (4\pi \sin \theta) / \lambda$, and 2θ is a scattering angle of the peak. The lamella thickness (l_c) was calculated from equation (3) on the basis of the degree of crystallinity (X_c) and the lamella periodicity (l_e) values.

$$l_c = (X_c/100) l_e \quad (3)$$

Fractured surface observation

Scanning Electron Microscope (SEM) observation of the fractured surface was performed on a Hitachi S-4700 (Japan) at room temperature with an acceleration voltage of 5 kV. The fractured surfaces of the

specimens were ion-sputtered with platinum and palladium in a vacuum.

RESULTS AND DISCUSSION

Characterization of polyacetal polymers

Polyacetal copolymers and polyacetal terpolymers compositions were determined by ¹H-MNR measurement, and the results are summarized in Table I. The polyacetal copolymers and the polyacetal terpolymers were obtained in 48–55% yields for the total monomers in feed. About 80% DOX and EHGE amounts in feed were incorporated into the polymer, which is in good agreement with the results reported in the previous article.¹⁷ During cationic ring-opening terpolymerization of TOX, DOX, and EHGE, chain transfer¹⁸ reaction may take place between cationic active sites and a hydroxyl end group in DMM. This reaction should affect the molecular weight of the polymers. Therefore, the M_w was controlled by a mount of DMM, that is, the usage of more amount of DMM leads to the lower molecular weight polymer. In the NMR spectra of the obtained polyacetal copolymers and polyacetal terpolymers, no signals assigned to homo-sequence of DOX unit and homo-sequence of EHGE unit were observed, indicating that the DOX and EHG units are randomly incorporated into the polymer backbone.¹⁹ The molecular weights of the polyacetal polymers were estimated by SEC measurements, and the results are summarized in Table I. The weight-average molecular weight (M_w) was controlled in amounts of both DMM as a chain transfer agent and BF₃ as a catalyst to research the influence on the creep behavior. The polyacetal copolymers and the polyacetal terpolymers having almost same

TABLE II
Solid Structures of Polyacetal Polymers

Polymers	l_e /nm	l_c /nm	X_c /%
Copolymer			
Cop-1	15	10	62
Cop-2	15	10	65
Cop-3	15	9	56
Terpolymer			
Ter-1	15	10	60
Ter-2	15	10	62
Homopolymer			
100P	17	13	69
500P	17	14	75

TABLE III
Tensile Properties and Activation Volumes of Polyacetal Polymers

Polyacetal polymers	Tensile strength/MPa	Tensile elongation/%	Activation volume/nm ³
Copolymer			
Cop-1	60.5	75	8.74
Cop-2	61.1	68	5.65
Cop-3	60.2	71	5.34
Terpolymer			
Ter-1	60.4	63	11.73
Ter-2	60.7	55	8.93
Homopolymer			
100P	69.0	65	4.16
500P	69.3	45	3.39

molecular weights to the polyacetal homopolymers 100P and 500P were chosen from the several polymerization samples and shown in Table I.

The solid state structures of the test piece in 1 mm thickness film were investigated by SAXS and the density value of the test piece. Volume fraction of the crystalline region (X_c), lamella periodicity (l_e), and lamella thickness (l_c) obtained by the crystal structure analysis are summarized in Table II. The both l_e and l_c values of the polyacetal copolymers and the polyacetal terpolymers are smaller than those of the polyacetal homopolymers.¹⁶ Those of the polyacetal copolymers and the polyacetal terpolymers does not depend on the difference of the molecular structure.

The tensile strength and the elongation are shown in Table III. Moreover, the plots of the degree of crystallinity (X_c) versus the tensile strength are shown in Figure 1. However, the tensile strength does not decrease linearly with a decrease in the crystallinity among the polyacetal polymers with three different structures, the polyacetal homopolymer, the polyacetal copolymers, and the polyacetal terpolymers. The tensile strength and crystallinity of the polyacetal homopolymers are higher than those of the polyacetal copolymers and the polyacetal terpolymers. It is, therefore, likely that the rearrangement of the crystals occurring at the deformation in creep or at yielding might be influenced by the difference in the lamella between the polyacetal homopolymer and the polyacetal copolymer.²⁰ Tashiro et al.²¹ investigated the structural change in the non-isothermal crystallization process of the polyacetal cooled from the molten state by the small-angle X-ray scattering (SAXS) and wide-angle X-ray scattering (WAXS) measurements as functions of temperature, and proposed a lamella insertion model of the polyacetal copolymer, where the amorphous region sandwiched between the neighboring two long lamella changes to a new lamella to form the stacked lamella structure with a shorter period.

Basic idea of creep rupture mechanism

The creep behavior is generally known to be analyzed by the deformation behavior and the rupture behavior.¹⁵ As an analysis method of the deformation behavior, viscoelastic studies have been carried out.²² However, the viscoelasticity analysis is limited to the amorphous polymers. While, for the crystalline polymers the linear strain region is limited to the initial deformation and the large deformation such as the creep strain, yielding or necking, indicates the nonlinear viscoelastic behavior because of unrecoverable structure change, due to the conformational change and rearrangement of the crystals. Especially, highly crystalline polyacetal homopolymer is considered to exhibit nonlinear viscoelastic deformation. It is well-known from the detailed studies based on nylons that the creep deformation curves on the various stresses can be superimposed by using a scale factor as the stress function instead of the stress and by subtracting the momentary

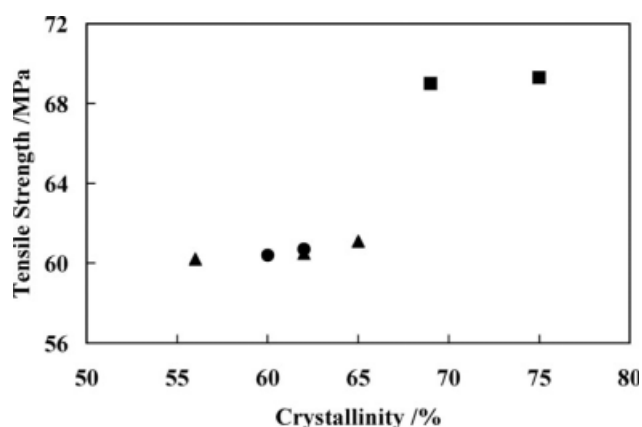


Figure 1 Plots of tensile strength versus crystallinity for (■) the polyacetal homopolymers, (▲) the polyacetal copolymers, and (●) the polyacetal terpolymers.

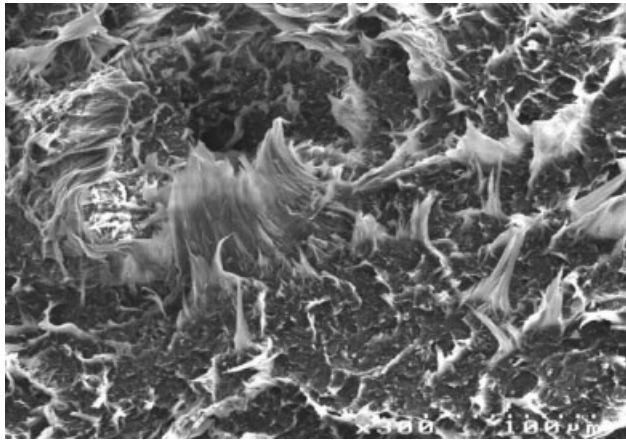


Figure 2 SEM micrograph of the fractured surface of creep rupture at 80°C.

elastic deformation.²³ It is considered as nonlinear viscoelasticity that the creep rupture behavior in the large deformation region for the polyacetal polymers can be treated as thermodynamics, same as yielding behavior.

The kinetics of the rupture process proposed by Zhurkov is generally applied to the analysis of the fracture behavior as the following equations.^{24–27} The kinetic concept of the fracture of the solids was proposed by Zhurkov and, therefore, the creep rupture time t_b is expressed by equation (4):

$$t_b = t_0 \exp[(\Delta F - v_c \sigma)/kT] \quad (4)$$

$$t_0 = h/(kT)$$

where ΔF , v_c , σ , k , h , and T are the activation energy, the activation volume, the loading stress, Boltzmann's constant, Planck's constant, and the absolute temperature, respectively. eq. (4) is rewritten by the logarithm:

$$\log t_b = \log t_0 + (\Delta F - v_c \sigma)/kT \quad (5)$$

This equation can be rewritten for the loading stress, σ :

$$\sigma = -2.303 k(T \log_{10} t_b)/v_c + \alpha \quad (6)$$

α is constant. From the experimental dependence, plotted as σ versus $\log t_b$, the kinetic parameters v_c and ΔF , which are characteristic values of materials, are determined.

Creep rupture behavior

It is, generally, well-known that the creep rupture mechanism of polymer may change from the ductile fracture to the brittle fracture in a low load region even if the creep rupture is ductile fracture in a high

load region.²⁵ To judge the fracture mechanism which it is the ductile fracture or the brittle one, the rupture surfaces of the creep test pieces were observed by SEM. As the example, the SEM micrograph of the rupture surface for polyacetal copolymer (Cop-2) on 21 MPa is shown in Figure 2, where the ruggedness is observed on the surface. Roughness on the rupture surface and the observation of the fibroid edge indicate that the rupture is due to the yielding, and that the creep fracture of polyacetal polymers is not brittle fracture but ductile fracture at the load of 20–30 MPa under the present experimental conditions.^{28,29} Therefore, the creep experiment was performed in the region where this ductile fracture is observed.

Plots of the creep rupture time, t_b , versus loading stress, σ , for the polyacetal homopolymers (100P and 500P) are shown in Figure 3, where longer rupture times are observed at lower loading stress for both 100P and 500P. However, the rupture time dependence on σ between the 100P and the 500P is different, and the slope for the 100P with high molecular weight is larger than that for the 500P with low molecular weight.³⁰ The activation volumes, v_c , can be calculated from the values which obtained from the slopes in Figure 3 correspond to the $-2.303 kT/v_c$ in equation (5), and summarized in Table III. The v_c value for the 100P with high molecular weight is to be 4.16, which is a little bit larger than that (3.39) for the 500P.

Plots of the creep rupture time, t_b , vs loading stress, σ , for the polyacetal copolymers (Cop-1, Cop-2, and Cop-3) are shown in Figure 4, and the rupture time dependence on loading stress for the Cop-1 with higher molecular weight is larger than that for the Cop-2 with lower molecular weight as well as the polyacetal homopolymers in Figure 3. And also, the values of the activation volume for the Cop-1, Cop-2, and Cop-3 are calculated from slopes

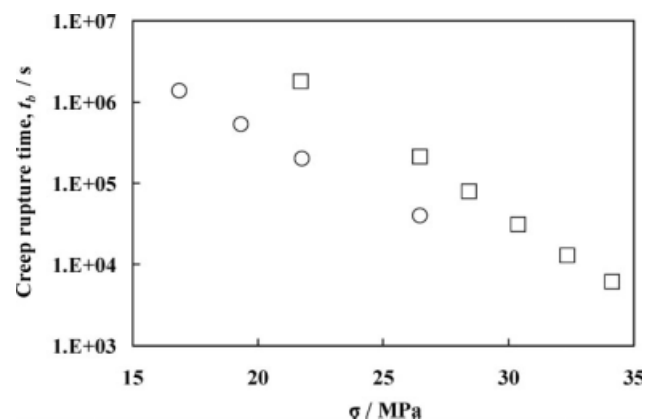


Figure 3 The relationship of creep rupture time (t_b) with loading stress (σ) for the polyacetal homopolymers of (□) 100P, and (○) 500P at 80°C.

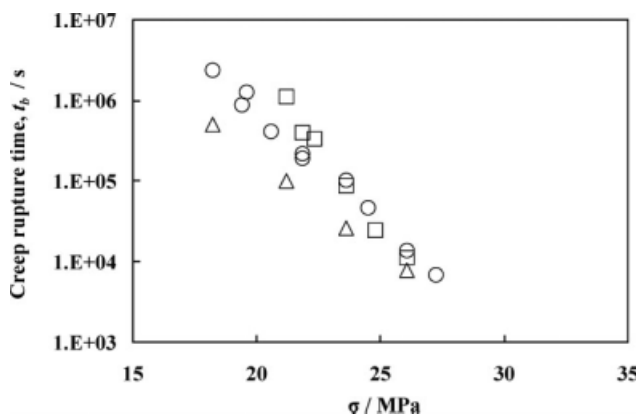


Figure 4 The relationship of creep rupture time (t_b) with loading stress (σ) for the polyacetal copolymers of (□) Cop-1, (○) Cop-2, and (Δ) Cop-3 at 80°C.

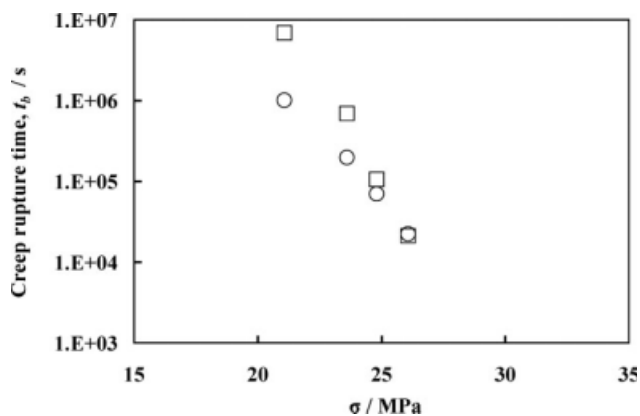


Figure 5 The relationship of creep rupture time (t_b) with loading stress (σ) for the polyacetal terpolymers of (□) Ter-1 and (○) Ter-2 at 80°C.

in the plots and the equation (5), and summarized in Table III. The value of the activation volume for the Cop-1 is larger than that for the Cop-2. Next, we investigated the effect of the comonomer content on the activation volume for Cop-2 and Cop-3. The rupture time dependence on loading stress are almost same for both polyacetal copolymers though the creep rupture time for the Cop-3 with high comonomer content is smaller than that for the Cop-2 with low comonomer content in the all region of the measurement. This indicates that the difference in 4–5 mol % of comonomer content in the polyacetal copolymers does not significantly affect the values of the activation volume. The polymer of the higher tensile yielding stress is inclined to have the longer creep rupture time.³¹ It is considered that shorter rupture time observed for the Cop-3 in comparison with Cop-2 is ascribed to a decrease in the yielding stress, exhibited by the tensile strength.

Plots of the creep rupture time, t_b , vs loading stress, σ , for the polyacetal terpolymers (Ter-1 and Ter-2) are shown in Figure 5, and the rupture time dependence on loading stress for the Ter-1 with higher molecular weight is larger than that for the Ter-2 with lower molecular weight, and also the values of the activation volume for both polyacetal terpolymers are summarized in Table III. Ter-1 has a larger v_c value than Ter-2. From the comparison in the activation volumes for all polyacetal polymers in Table III, the polyacetal polymers with higher molecular weight have larger values of the activation volume than those with lower molecular weight, and also polyacetal terpolymers have larger values of the activation volume than polyacetal homopolymer and polyacetal copolymers.

As the activation volume is dependent upon the molecular weight, the effect of the molecular weight on it was investigated. The relationship of the values of the v_c with the molecular weights of the polyacetal

polymers is shown in Figure 6. When the activation volume were compared among the polyacetal polymers with the same molecular weights, they increase in the following order: polyacetal homopolymers < the polyacetal copolymers < polyacetal terpolymers. This finding is in good agreements with the result that the occurrence rate of the crack on the fatigue experiments is slower in the polyacetal copolymers compared with the polyacetal homopolymers.³² It is reported that although both polyacetal polymers have the same crystal structures, their lamella structures in the crystals are different.^{33,34} It is presumed that the difference of the activation volume between the polyacetal polymers is caused by the physical crosslinking structure of the both neighboring lamellae. It is likely that the rearrangement of the crystals occurring at the deformation in creep might be influenced by the difference in the comonomer structure. The reason why the polyacetal terpolymers have higher activation volume than the

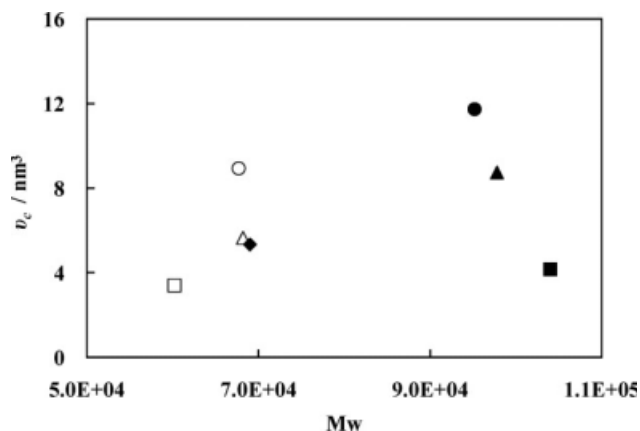


Figure 6 Plots of creep activation volume versus molecular weights (M_w) for the polyacetal homopolymers [(■) 100P and (□) 500P], the polyacetal copolymers [(▲) Cop-1, (Δ) Cop-2, and (◆) Cop-3], and the polyacetal terpolymers [(●) Ter-1 and (○) Ter-2], respectively.

polyacetal copolymers is not clear at present. However, it is reported that the branched polyacetal polymers has good tensile strength properties and the modulus like the shih-kebab morphology.¹² Although the polyacetal terpolymers and the polyacetal copolymers in this work have the almost same mechanical properties and the morphology, it is likely that the short branched structures present in the polyacetal terpolymers, due to the EHGE unit as the comonomer component, may provide the high yielding strength³⁵ to the polyacetal terpolymers in the creep process as the slow strain property.

CONCLUSIONS

Polyacetal copolymers were prepared by cationic ring-opening copolymerizations of 1,3,5-trioxane (TOX) with 1,3-dioxolane (DOX), and polyacetal terpolymers were prepared by terpolymerizations of TOX, DOX, and 2-ethylhexyl glycidyl ether (EHGE). The polymers were obtained in 48–50% yields, and about 80% of DOX and EHGE amounts in feed were incorporated in their polymers as the comonomer units.

From SEM observation, this creep rupture behavior can be treated as ductile behavior, and the kinetics of the rupture process proposed by Zhurkov is generally applicable to the analysis of the creep rupture behavior. Creep rupture was characterized by the activation volume, v_c , value in Zhurkov's equation, which can be estimated from the slope in the plots of load versus log (rupture time) at 80°C. The polyacetal polymers with higher molecular weight have larger values of the activation volume than those with lower molecular weight. When the activation volume values are compared among the polyacetal polymers with the same molecular weights, they increase in the following order: the polyacetal homopolymers < the polyacetal copolymers < polyacetal terpolymers.

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