

# Dielectric Properties above the Glass Transition for a Series of Epoxide Prepolymers

TSUNEO KOIKE\* and RYOHEI TANAKA

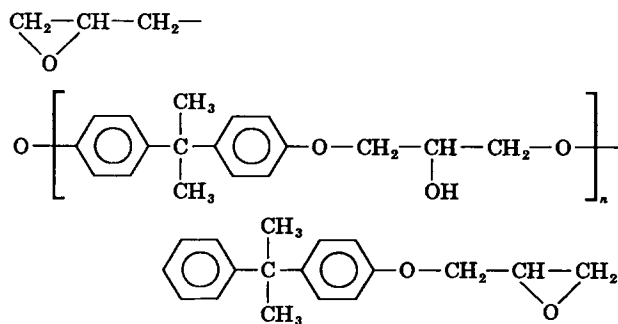
Research and Development Laboratory, Yuka Shell Epoxy Co., Ltd., Shiohama-cho 1, Yokkaichi-city, Mie, 510, Japan

## SYNOPSIS

Dielectric properties above the glass transition have been investigated for a series of bisphenol-A type epoxide prepolymers ( $388 \leq \bar{M}_w \leq 2640$ ). Dielectric measurements were performed over a frequency range of 50 Hz–1 MHz using a vertical parallel plate cell which was constructed in the laboratory. The dielectric  $\alpha$ -relaxation for each prepolymer fits the empirical model of the Havriliak–Negami equation. The temperature dependence of the dielectric relaxation time  $\tau$  is described by the Williams–Landel–Ferry (WLF) equation as well as that of the direct current conductivity  $\sigma$ , which can be measured using the same cell. The relationship between  $\tau$  and  $\sigma$ ,  $\sigma \cdot \tau^m = \text{const}$ , is derived from experimental results. The exponent  $m$ , which depends on the molecular weight of the prepolymer, is considered to correspond to the ratio of the segmental mobility to ionic mobility. The dielectric loss  $\epsilon''$  can be used as an indicator of the direct current conduction in the temperatures where the ionic component in  $\epsilon''$  becomes much larger than the dipole one.

## INTRODUCTION

Dielectric analysis is widely used to monitor the curing process of epoxy resin system from the stand point of nondestructive inspection and automatic measurement.<sup>1–5</sup> In spite of many efforts to analyze the curing mechanism by dielectric measurements, the mechanism is not always clarified because of a variety of effects caused by the epoxide prepolymer, the hardner, and their reactants, which have a wide range of molecular weights.<sup>6</sup> The most popular epoxide prepolymer is the diglycidylether of bisphenol A (DGEBA) whose structural formula is shown below<sup>7</sup>:



\* To whom correspondence should be addressed.

The epoxide prepolymer has different molecular weights according to the variation of the repeated number  $n$  indicated above. A study on the dielectric properties of a series of prepolymers, which have different molecular weights, may provide useful information for the detailed analysis of the epoxy resin curing process.

The dielectric properties of an epoxide prepolymer have been measured below the glass transition temperature ( $T_g$ ) and have been discussed on  $\beta$ - and  $\gamma$ -relaxation by Pochan and co-workers.<sup>8</sup> Usually, an epoxide prepolymer is cured in a liquid state above  $T_g$  after mixing with suitable hardners, so that the study on the  $\alpha$ -relaxation may be necessary for a better understanding of the curing mechanism. An epoxide prepolymer changes from an amorphous solid to a viscous liquid state<sup>1,9,10</sup> in the temperature range of  $T_g$  to about  $T_g + 100^\circ\text{C}$ . Therefore, it is not easy to perform the dielectric measurements of the prepolymer above  $T_g$ , by using the commercially available specimen holder which has been applied to the measurement below  $T_g$ .<sup>8</sup> In previous work,<sup>11</sup> we have carried out the dielectric measurements of the curing process for an epoxy resin system by using a simple design measuring cell developed in our laboratory. The cell can be used for the dielectric measurement of the epoxide prepolymer in a solid to a liquid state as well as during the curing process.

Dielectric relaxation time  $\tau$  can be obtained by the Cole-Cole plot<sup>12,13</sup> in the  $\alpha$ -relaxation region. Sasabe and Saito<sup>14</sup> have reported the empirical equation between  $\tau$  and direct current (dc) conductivity  $\sigma$ ,  $\sigma \cdot \tau^m = \text{const}$ , for several amorphous polymers: poly(vinyl chloride), poly(vinylidene chloride), poly(vinyl acetate), amorphous poly(ethylene terephthalate), and polycarbonate. The equation corresponds to Walden's rule<sup>15</sup> between  $\sigma$  and viscosity  $\eta$ ,  $\sigma \cdot \eta = \text{const}$ , for low molecular weight liquids, and also to a modified Walden's rule,  $\sigma \cdot \eta^m = \text{const}$ , reported by Sazhin and Shuvayev<sup>16</sup> for polystyrene solutions. The equation of this type is not yet applied to amorphous oligomers such as epoxide prepolymers.

The objective of this study is to clarify the dielectric properties of epoxide prepolymers above  $T_g$ , and to correlate them to the dc conductivity, which is also used for the measurement of the epoxy resin curing process due to the close association with the viscoelastic properties of epoxy resin systems.<sup>17,18</sup>

## EXPERIMENTAL

### Materials

The epoxide prepolymers used in this study were five oligomers of the diglycidylether of bisphenol-A with different weight-average molecular weights ( $388 \leq \bar{M}_w \leq 2640$ ). The five epoxide prepolymers, Epikote 828, 834, 1001, 1002, and 1004, were manufactured by a conventional process<sup>7</sup> in Yuka Shell Epoxy. The characteristics of the samples are given in Table I. Number-average molecular weights ( $\bar{M}_n$ ) of the samples were measured in a vapor pressure osmometer (VPO-117; Corona Electronics) at 57°C with methyl ethyl ketone (MEK) as the solvent. Weight-average molecular weights ( $\bar{M}_w$ ) were computed from the results obtained by a gel permeation chromatograph (Model 600; Waters Associates)

with a column system, TSK G2500 + G3000H (Toyo Soda). The column system was calibrated by polystyrene samples according to the method of Mori.<sup>19</sup> The glass transition temperature was measured by duPont 9000 differential scanning calorimeter.<sup>20</sup>

### Measurements

Dielectric measurements were performed using an inductive-ratio-arm bridge type apparatus (TR-1100; Ando Electric). The measuring frequency was 50 Hz–1 MHz, in the temperature range of 0–180°C. A vertical parallel plate cell, based on the three-terminal method, was used to measure the samples (Fig. 1). Temperature corrections for the geometric capacitance of the cell were made on all measurements in order to eliminate errors caused by the thermal expansion of the cell. The cell was improved from the one which was developed in previous work,<sup>11</sup> in the following points:

- adopting glass plates instead of polycarbonate resin sheet for the wall of the cell;
- using gold-plated copper foil for the electrodes in order to avoid the corrosion of copper.

This type of cell makes it possible to measure the sample not only in the liquid but also in the solid state because of the vertical design and simple shape of the cell.<sup>11</sup> Accurate measuring data can be obtained by using this three-terminal cell in comparison with other vertical type cells, based on the two-terminal (without guard electrode) method, developed by Delmonte<sup>1</sup> and Lane.<sup>3</sup>

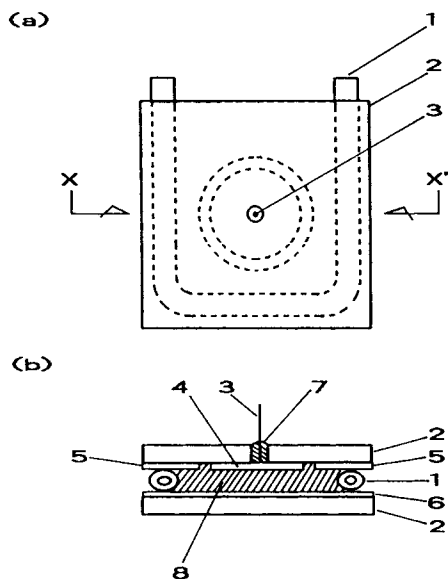
The dc conductivity measurements were performed in the temperature range of 0–180°C using an apparatus system which consisted of a dc power supply (TR300C; Takeda Riken), a vibrating reed electrometer (TR-84M; Takeda Riken), and a pen recorder (EPR-200A; Toa Dempa). The measuring

**Table I** Properties of Epoxide Prepolymers

Prepolymer	$T_g$ (°C)	$\bar{M}_n$	$\bar{M}_w$	$\bar{M}_w/\bar{M}_n$	Na <sup>+</sup> <sup>a</sup> (ppm)	Cl <sup>-</sup> <sup>b</sup> (ppm)
Epikote 828	-16	366	388	1.06	< 1	< 1
Epikote 834	0	479	590	1.23	1	< 1
Epikote 1001	30	898	1396	1.55	< 1	< 1
Epikote 1002	39	1147	1891	1.63	1	< 1
Epikote 1004	54	1538	2640	1.72	2	< 1

<sup>a</sup> Measured by atomic absorption analysis.

<sup>b</sup> Measured by potentiometric titration with 0.01 N silver nitrate.



**Figure 1** Configuration of a vertical parallel plate cell. (a) Side view; (b) cross section view ( $X - X'$ ): (1) silicone tube (outer diameter, 4 mm), (2) glass plate (length  $\times$  width  $\times$  thickness, 50  $\times$  50  $\times$  3 mm) (3) copper wire (diameter, 1 mm), (4) electrode (gold-plated copper foil; diameter, 20 mm; thickness, 0.035 mm), (5) guard electrode (gold-plated copper foil; inner diameter, 22 mm; thickness, 0.035 mm), (6) electrode (gold-plated copper foil; length  $\times$  width  $\times$  thickness, 50  $\times$  50  $\times$  0.035 mm), (7) epoxy adhesive, (8) epoxide prepolymer (thickness, 1.5–2.5 mm).

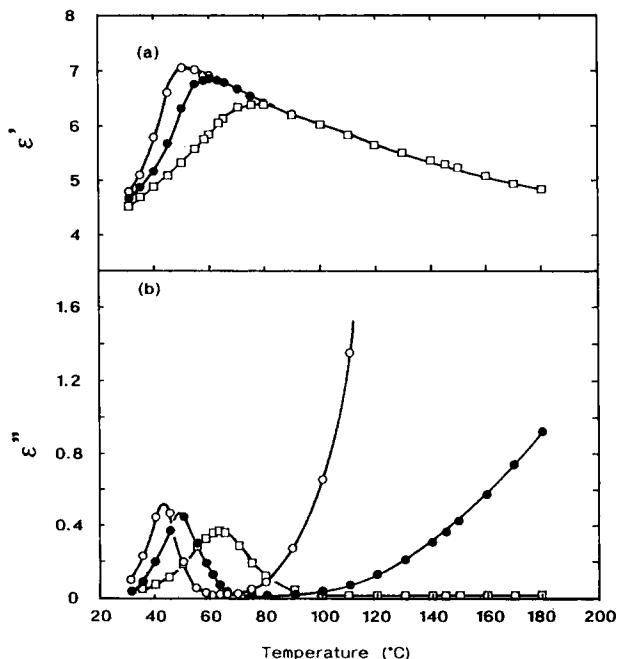
cell was the same one used for the dielectric measurements.

## RESULTS AND DISCUSSION

### Dielectric Properties

Figure 2 shows the dielectric behavior of an epoxide prepolymer, Epikote 1001, for three frequencies (a total of 10 were measured) over a temperature range of 30–180°C. A single relaxation process was observed above  $T_g$  for each frequency. The dielectric loss  $\epsilon''$  of Epikote 1001 was plotted as a function of frequency at the temperatures in the relaxation region (Fig. 4). A broad  $\alpha$ -relaxation was observed at each temperature, which was due to the micro-Brownian motions of the amorphous prepolymer chain.<sup>21,22</sup> Similar results were obtained for Epikote 1004, as shown in Figures 3 and 5, and for other three prepolymers.

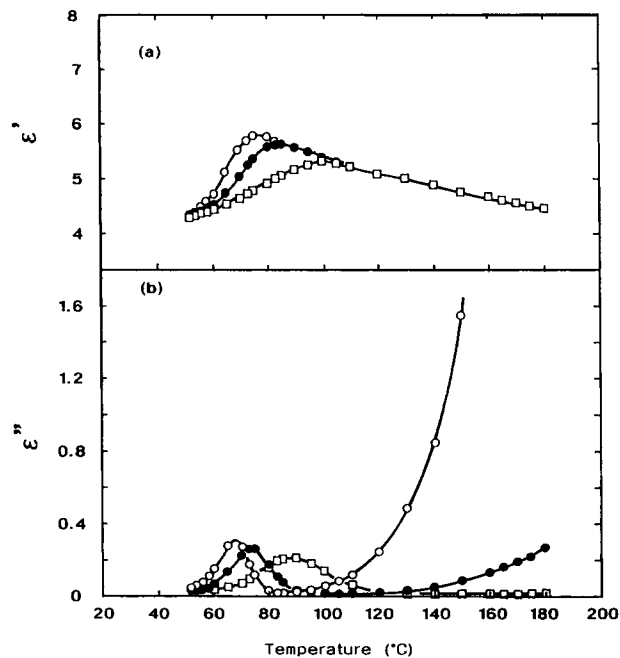
The sharp increase of dielectric loss, caused by the ionic conduction,<sup>21</sup> can be seen at higher temperatures in Figures 2 and 3. This behavior will be



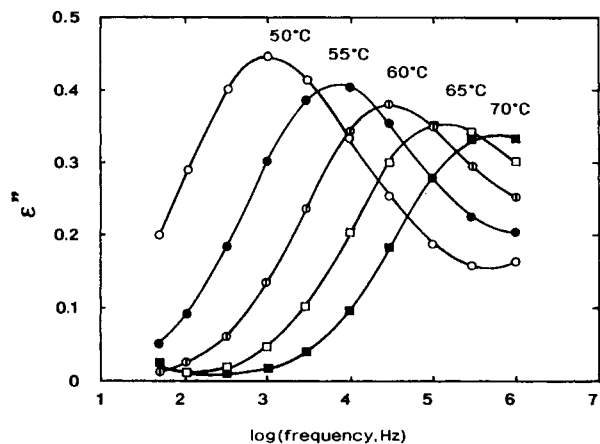
**Figure 2** Temperature dependence of (a)  $\epsilon'$  and (b)  $\epsilon''$  for Epikote 1001 at three frequencies: (○) 50 Hz; (●) 1 kHz; (□) 100 kHz.

discussed later comparing directly with the result of the dc conduction measurements.

In order to define the relaxation process, the Cole-



**Figure 3** Temperature dependence of (a)  $\epsilon'$  and (b)  $\epsilon''$  for Epikote 1004 at three frequencies; (○) 50 Hz; (●) 1 kHz; (□) 100 kHz.



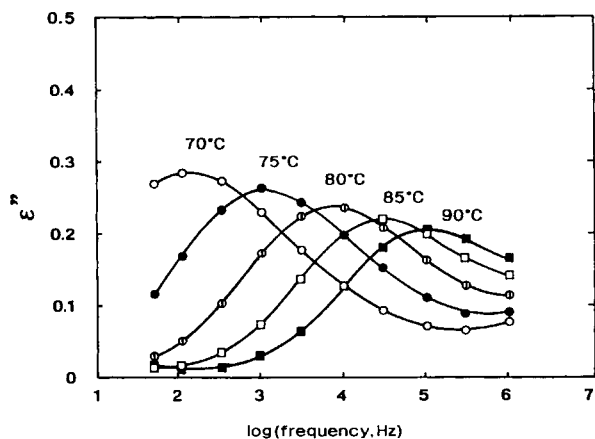
**Figure 4** Frequency dependence of  $\epsilon''$  at various temperatures for Epikote 1001 in the  $\alpha$ -relaxation region.

Cole plots for the prepolymers were studied. The following empirical model of Havriliak and Negami<sup>23</sup> is applicable to the dielectric relaxation process for most polymers:

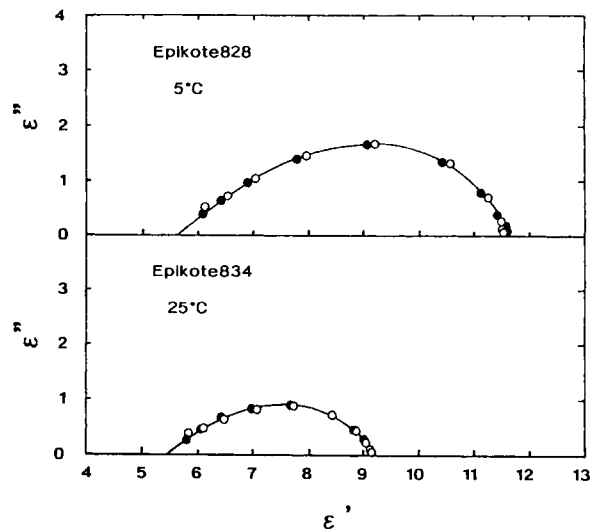
$$\epsilon^*(\omega) - \epsilon_\infty = \frac{\epsilon_0 - \epsilon_\infty}{[1 + (i\omega\tau)^{1-\alpha}]^\beta} \quad (1)$$

where  $\epsilon^*(\omega)$  = the complex dielectric constant,  $\epsilon_\infty$  = high-frequency limit to the dielectric constant,  $\epsilon_0$  = low-frequency limit to the dielectric constant,  $\omega$  = angular frequency,  $\tau$  = relaxation time,  $\alpha$  = distribution parameter, and  $\beta$  = skewness parameter.

Figures 6 and 7 show Cole-Cole plots for five prepolymers, comparing experimental values with calculated ones. These calculated values fitted well experimental ones for five prepolymers. Table II shows the best fit parameters obtained according to eq. (1).

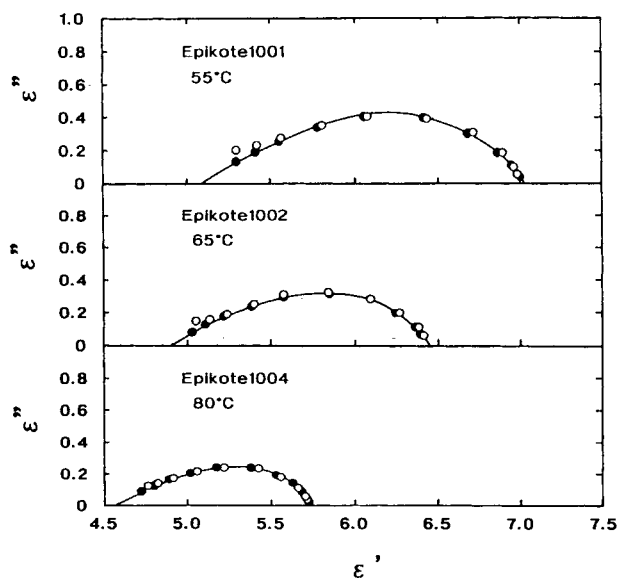


**Figure 5** Frequency dependence of  $\epsilon''$  at various temperatures for Epikote 1004 in the  $\alpha$ -relaxation region.



**Figure 6** Cole-Cole arcs for Epikote 828 at 5°C and Epikote 834 at 25°C (○) experimental value; (●) calculated value.

The parameters in Table II can be associated with the properties of the prepolymers as well as those of polymers in Havriliak and Negami's work.<sup>23</sup> The value  $\epsilon_0 - \epsilon_\infty$  represents the effective moment of the orienting unit, so that the lower  $\bar{M}_w$  prepolymer has the larger value. Two parameters  $\alpha$  and  $\beta$  slightly change within the temperature range examined. The parameter  $\alpha$  described the width of the dispersion; the width increases as  $\alpha$  ranges from zero to unity.



**Figure 7** Cole-Cole arcs for Epikote 1001 at 55°C, Epikote 1002 at 65°C, and Epikote 1004 at 80°C: (○) experimental value; (●) calculated value.

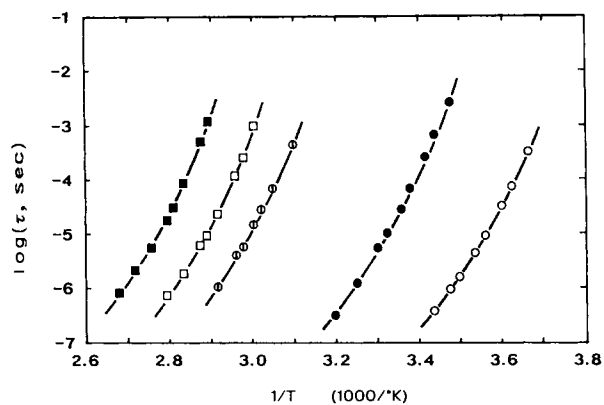
**Table II** Parameters of the Equation of Havriliak–Negami for Epikote Prepolymers

Prepolymer	Temp (°C)	$\epsilon_0$	$\epsilon_\infty$	$\epsilon_0 - \epsilon_\infty$	$\alpha$	$\beta$	Relaxation Time (s)
Epikote 828	0	12.00	5.55	6.45	0.24	0.59	$3.2 \times 10^{-4}$
	5	11.62	5.68	5.94	0.24	0.59	$3.0 \times 10^{-5}$
	10	11.29	5.77	5.52	0.25	0.60	$4.3 \times 10^{-6}$
Epikote 834	20	9.43	5.25	4.18	0.32	0.56	$2.5 \times 10^{-4}$
	25	9.16	5.37	3.79	0.33	0.57	$2.7 \times 10^{-5}$
	30	8.93	5.38	3.55	0.33	0.57	$5.7 \times 10^{-6}$
Epikote 1001	50	7.14	5.01	2.13	0.34	0.50	$4.2 \times 10^{-4}$
	55	7.02	5.06	1.96	0.35	0.51	$7.0 \times 10^{-5}$
	60	6.91	5.10	1.81	0.35	0.51	$1.5 \times 10^{-5}$
Epikote 1002	60	6.57	4.84	1.73	0.36	0.50	$8.6 \times 10^{-4}$
	65	6.44	4.87	1.57	0.36	0.50	$1.1 \times 10^{-4}$
	70	6.33	4.88	1.45	0.36	0.50	$2.4 \times 10^{-5}$
Epikote 1004	75	5.83	4.55	1.28	0.37	0.46	$5.0 \times 10^{-4}$
	80	5.74	4.55	1.19	0.37	0.46	$8.3 \times 10^{-5}$
	85	5.65	4.55	1.10	0.38	0.47	$1.8 \times 10^{-5}$

The prepolymer with narrower molecular weight distribution, Epikote 828, has smaller value of  $\alpha$  than other prepolymers. The parameter  $\beta$  describes the skewness of the dispersion; the skewness increases as  $\beta$  ranges from unity to zero. Epikote 1004, which has higher  $M_w$  and wider distribution, has larger skewness of the dispersion according to the small value of  $\beta$ .

#### Relationship between Dielectric Relaxation Time and dc Conductivity

Figures 8 and 9 show the temperature dependence of  $\tau$  and the dc conductivity  $\sigma$ , respectively, for five prepolymers. Figure 10 shows both the temperature

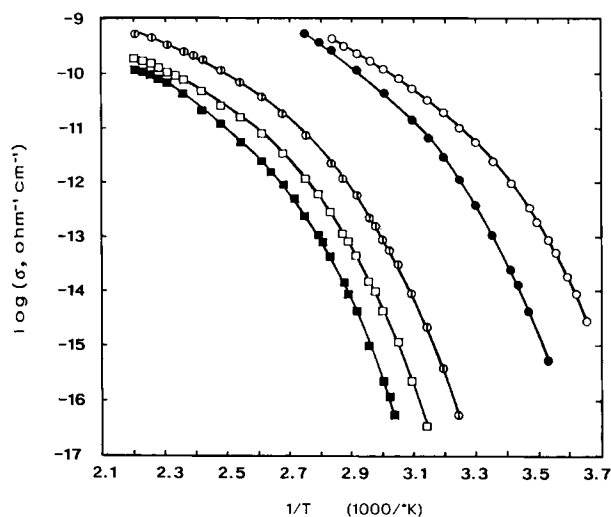


**Figure 8** Temperature dependence of relaxation time for five prepolymers: (○) Epikote 828; (●) Epikote 834; (○) Epikote 1001; (□) Epikote 1002; (■) Epikote 1004.

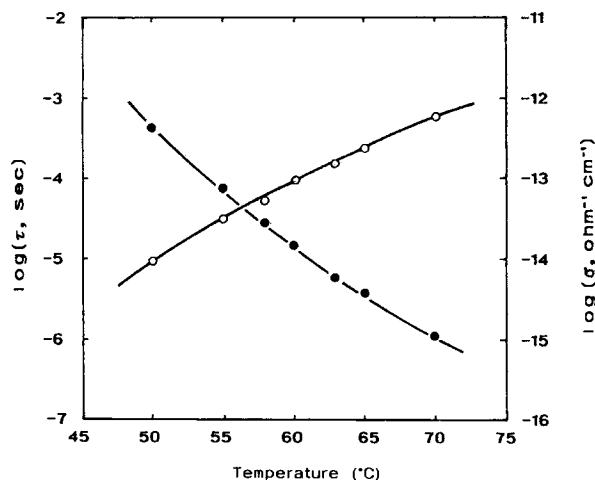
dependence of  $\tau$  and that of  $\sigma$  for Epikote 1001. The relation between  $\tau$  and  $\sigma$  is shown for five prepolymers by the log–log plot in Figure 11. This relation can be described in the following equation from the linear plots in Figure 11.

$$\sigma(T)[\tau(T)]^m = \text{const} \quad (2)$$

where  $T$  is temperature and  $m$  is exponent. The value of the exponent  $m$  varies from 0.62 to 0.82 according to  $M_w$  of the prepolymers as shown in Table III.



**Figure 9** Temperature dependence of dc conductivity for five prepolymers: (○) Epikote 828; (●) Epikote 834; (○) Epikote 1001; (□) Epikote 1002; (■) Epikote 1004.



**Figure 10** Temperature dependence of relaxation time and dc conductivity for Epikote 1001: (●) relaxation time; (○) dc conductivity.

In order to understand the meaning of  $m$ , the behavior of both  $\tau$  and  $\sigma$  was analyzed. It is generally recognized that the temperature dependence of  $\tau$  above  $T_g$  is governed by the WLF equation<sup>24</sup>

$$\log \frac{\tau(T)}{\tau(T_g)} = \frac{-C'_1(T - T_g)}{C'_2 + T - T_g} \quad (3)$$

where  $T$  is temperature,  $T_g$  is glass transition temperature, and  $C'_1$  and  $C'_2$  are parameters.

On the other hand, the dc conduction for polymers above  $T_g$  is considered to be caused by ions<sup>25</sup> (their mobility and concentration), which are always present, to some extent, in commercial products. The major ions in epoxide prepolymers are  $\text{Na}^+$  and  $\text{Cl}^-$  due to the manufacturing process.<sup>7,26</sup> Table I shows that the ion concentrations are regarded as similar among the prepolymers examined. Therefore, the dc conductivity in the prepolymer is mainly affected by the ionic mobility, which is closely related to the segmental mobility of the host prepolymer. The WLF equation has been applied to the temperature dependence of  $\sigma$  above  $T_g$  for several polymers by Saito and co-workers.<sup>25</sup>

$$\log \frac{\sigma(T)}{\sigma(T_g)} = \frac{C''_1(T - T_g)}{C''_2 + T - T_g} \quad (4)$$

where  $C''_1$  and  $C''_2$  are parameters. Miyamoto and Shibayama<sup>27</sup> have reported that the dc conduction for three types of crosslinked epoxy resins was caused by ions, and the temperature dependence of  $\sigma$  above  $T_g$  was governed by the WLF equation.

As for epoxide prepolymers, the temperature de-

pendence of  $\tau$  and  $\sigma$  was also found to fit the WLF equation. Table III shows the parameters,  $C'_1$ ,  $C'_2$ ,  $C''_1$ , and  $C''_2$ , compared to the value of  $m$ . The following relations are obtained for each prepolymer from the result in Table III:

$$C'_1 \neq C''_1 \quad (5)$$

$$C'_2 \approx C''_2 \quad (6)$$

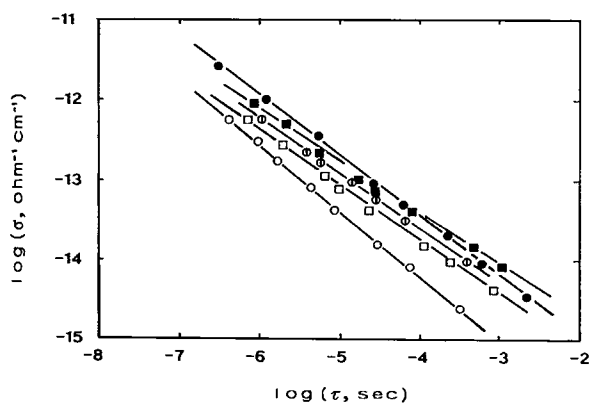
$$m \approx C''_1/C'_1 \quad (7)$$

These relations agreed with the result of Sasabe's work on polymers.<sup>14</sup> Sasabe has made an explanation on the value of  $m$ ;  $m$  corresponded to the ratio of the ionic mobility and the segmental one for the polymers according to both the WLF equation and the free volume theory developed by Cohen and Turnbull<sup>28,29</sup>:

$$C_1 = \frac{\gamma v^*}{2.303 f_g v_m} \quad \text{and} \quad C_2 = \frac{f_g}{\alpha_f} \quad (8)$$

where  $C_1$ ,  $C_2$  = WLF parameters,  $\gamma$  = a numerical factor introduced to correct for overlap of free volume,  $v^*$  = the critical volume large enough to permit a molecule to jump in after the displacement  $v_m$  = the average molecular volume,  $f_g$  = the free volume fraction at  $T_g$ , and  $\alpha_f$  = thermal expansion coefficient of the free volume.

The parameter  $C_2$ , as shown in eq. (8), reflects the characteristics of a polymer matrix or a host polymer matrix only. The parameter  $C_1$  is determined not only by the characteristics of a polymer (a host polymer) matrix, but also by the size of the moving unit, which is expressed by the critical volume  $v^*$  for the transport of the polymer segment or



**Figure 11** Relation between  $\log \tau$  and  $\log \sigma$  for five prepolymers: (○) Epikote 828; (●) Epikote 834; (◊) Epikote 1001; (□) Epikote 1002; (■) Epikote 1004.

**Table III** WLF Parameters for Epoxide Prepolymers

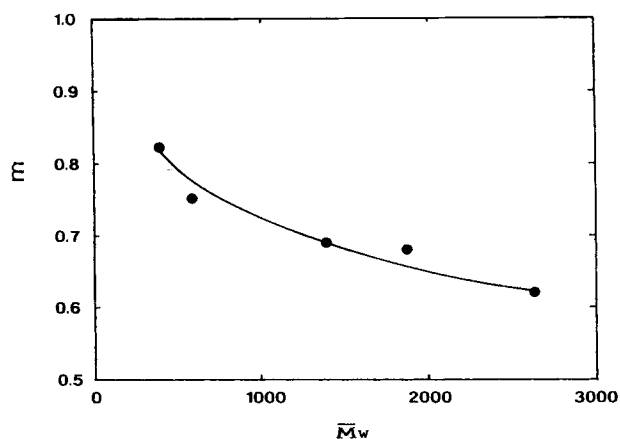
Prepolymer	$T_g$ (°C)	$C_1$	$C_2$	$C_1''$	$C_2''$	$C_1''/C_1$	$m$
Epikote 828	-16	15.69	28.0	12.92	28.0	0.82	0.82
Epikote 834	0	16.74	35.5	12.29	33.9	0.73	0.75
Epikote 1001	30	16.14	48.9	10.99	48.3	0.68	0.69
Epikote 1002	39	16.54	48.8	11.05	47.1	0.67	0.68
Epikote 1004	54	15.46	52.6	9.76	54.6	0.63	0.62

the ionic charge carrier. Consequently, the exponent  $m$  is regarded as a measure of the size of the ionic charge carrier in comparison to that of moving polymer segments.

The behavior of the epoxide prepolymer can be explained on the same concept. The molecular weight dependence of the exponent  $m$  for the epoxide prepolymers is shown in Figure 12. The value of  $m$  is less than unity for each prepolymer, which means that the critical volume for the ion transport is smaller than that for the prepolymer segment. The prepolymer with a larger  $\bar{M}_w$  has a smaller value of  $m$ . This behavior is interpreted in the following way. The five prepolymers are considered to have the same kinds of ions due to the same manufacturing process. The critical volume for the ion transport is not so different among the prepolymers while that for the segment transport increases with the increase of  $\bar{M}_w$ .

### Comparison of Dielectric Loss and dc Conductivity

The dielectric loss  $\epsilon''$  has a dipolar and an ionic component;



**Figure 12** Relation between  $m$  and  $\bar{M}_w$  for five prepolymers.

$$\epsilon'' = \epsilon_d'' + \epsilon_i'' \quad (9)$$

where  $\epsilon_d''$  is a dipolar component and  $\epsilon_i''$  is an ionic one.<sup>21</sup>

The ionic term  $\epsilon_i''$  is mainly attributed to the dc conduction, and can be described as follows<sup>21</sup>:

$$\epsilon_i'' = \sigma / \omega E_0 \quad (10)$$

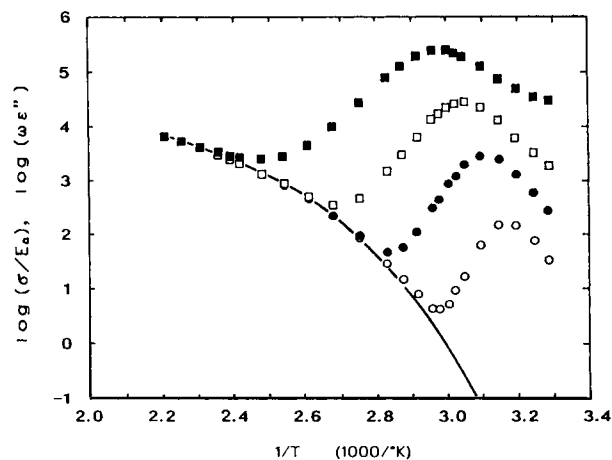
where  $E_0$  is the permittivity of free space ( $8.555 \times 10^{-14}$  F/cm),  $\sigma$  is dc conductivity, and  $\omega$  is angular frequency.

Equation (9) can be rearranged in the following form by combining with eq. (10):

$$\omega \epsilon'' = \omega \epsilon_d'' + \sigma / E_0 \quad (11)$$

The term  $\omega \epsilon_d''$  becomes relatively small due to the significant increase of  $\sigma$  in the temperatures where active segmental motions take place. If the value of the product  $\omega \epsilon_d''$  is negligible, the product  $\omega \epsilon''$  can be a direct measure of the dc conduction.<sup>21</sup>

The five prepolymers, as shown in Figure 9, have larger values of  $\sigma$  in high temperatures. Figure 13



**Figure 13** Temperature dependence of  $\sigma/E_0$  and  $\omega \epsilon''$  at four frequencies for Epikote 1001: (○) 50 Hz; (●) 1 kHz; (□) 10 kHz; (■) 100 kHz.

shows the temperature dependence of  $\omega\epsilon''$  at four frequencies for Epikote 1001 in comparison with that of  $\sigma/E_0$  calculated from the results of the dc conduction measurements. A broad peak is observed for four frequencies, respectively, in low temperatures on the plot of the product  $\omega\epsilon''$  versus the inverse of the absolute temperature  $1/T$ , which is due to the rotational diffusion of the dipole moments.<sup>22</sup> In high temperatures, data at the four frequencies superimposed onto one curve (solid line), which was made by the plot of  $\sigma/E_0$  versus  $1/T$ . The better agreement is observed at the lower frequency in Figure 13. Other prepolymers also exhibited the similar tendency. Consequently, the dielectric loss  $\epsilon''$  for the epoxide prepolymer can be used as an indicator of the dc conduction at a fixed frequency in the temperatures where the dipole component is negligible.

The result mentioned above and the empirical equation between  $\tau$  and  $\sigma$ ,  $\sigma \cdot \tau^m = \text{const}$ , may provide valuable useful information for a better understanding of the curing mechanism of epoxy resin in the dielectric cure monitoring.

## CONCLUSIONS

Dielectric properties above the glass transition have been investigated for a series of bisphenol-A type epoxide prepolymers ( $388 \leq \bar{M}_w \leq 2640$ ) by using a vertical parallel plate cell, in comparison with the dc conductivity. The following conclusions were obtained.

- (a) The dielectric  $\alpha$ -relaxation fits to the empirical model of Havriliak–Negami equation.
- (b) The temperature dependence of the dielectric relaxation time  $\tau$  above  $T_g$  is described by the WLF equation as well as that of the dc conductivity  $\sigma$ .
- (c) The relationship between  $\tau$  and  $\sigma$ ,  $\sigma \cdot \tau^m = \text{const}$ , is experimentally derived. The exponent  $m$ , which depends on the molecular weight of the prepolymer, is considered to be a measure of the ratio of the segmental mobility to ionic mobility.
- (d) The dielectric loss  $\epsilon''$  can be used as an indicator of the dc conduction in the temperatures where the ionic component in  $\epsilon''$  becomes much larger than the dipole one.

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