# Bonding Properties of Epoxy Resins Containing Two Mesogenic Groups

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**ABSTRACT:** Liquid-crystalline epoxy resins, with introduced aliphatic chains between two mesogenic groups, were synthesized and their adhesive bonding properties were compared to those of the bisphenol-A–type epoxy resin and the liquid-crystalline epoxy resin, previously reported. The bonding strength of the former resin system was higher than that of the two later systems. We suggest that the high bonding strength of the twin mesogenic epoxy resins, cured with an aromatic amine, was attributable to the large plastic deformation of the adhesive layer in the fracturing process. We also investigated the effects of the aliphatic chain length in the twin mesogenic epoxy resin on their dynamic mechanical and bonding properties. The bonding strength of the cured twin mesogenic epoxy resins increased with an increase in the aliphatic chain length. We suggest that the high bonding strength of the system introduced by the long aliphatic chain was attributable to the large plastic deformation of the adhesive layer because of the higher network mobility. © 2004 Wiley Periodicals, Inc. J Appl Polym Sci 92: 3721–3729, 2004

**Key words:** resins; adhesives; mechanical properties; fracture; liquid-crystalline epoxies

## INTRODUCTION

Epoxy resins are widely used as adhesives and coatings because of their many desirable properties, such as good bonding strength, excellent thermal and chemical stabilities, and electrical properties.[1] Even now, the development of high-performance epoxy resins is required in many industrial fields, for example, the machine, electric, and electronic industries.

Liquid-crystalline epoxy resins that contain mesogenic groups in their backbone, such as the biphenol-type,[2–5] azine-type,[6] stilbene-type,[7] and hydroxybenzoic acid–type resins[8,9] have been noted as novel high-performance epoxy resins. Possible applications of these epoxy resins include wave-guides and mechanically switched optical devices,[9] for instance. Many investigations have been reported on the synthesis and both optical and mechanical properties of the liquid-crystalline epoxy resins.[2–11] We also previously discussed the relationship between mechanical properties and network structure of the liquid-crystalline epoxy resins.[2,3,5] In these reports, we suggested that the liquid-crystalline epoxy resins cured with an aromatic amine show good toughness and bonding properties because of the large plastic deformation caused by the reorientation of mesogenic groups in the fracture process.

Generally speaking, the liquid-crystalline epoxy resins are usually solid at room temperature and their melting points are considerably higher than that of a common epoxy resin. Thus, the conditions under which liquid-crystalline epoxy resins are cured must be more severe. In addition, the internal stress occurring in the adhesive joints with the liquid-crystalline epoxy resins is usually higher than that of the common epoxy resin, given their high glass-transition temperature. In a previous study,[12] we investigated the bonding properties of the terephthalylidene-type liquid-crystalline epoxy resin, whose melting point was about 190°C. The internal stress of the cured resin was about three times higher than that of the common epoxy resin. Although the liquid-crystalline epoxy resins observed in a previous study showed a high bonding strength, the high melting point and high internal stress were their disadvantages.

Recently, Ober et al.[13] reported the synthesis and characterization of a twin mesogenic epoxy resin that has flexible aliphatic chains between two mesogenic groups. Choi et al.[14] also reported that the twin mesogenic resins, which have some long aliphatic chains [number ($n$) of carbon atoms > 6], have relatively low transition temperatures.

In this study, we synthesized some twin mesogenic epoxy resins that have flexible aliphatic chains in their backbone moiety to overcome the above disadvantages of the terephthalylidene-type epoxy resin. The bonding properties of these resins are discussed and compared with those of the terephthalylidene-type epoxy resin used in the previous study as a liquid-
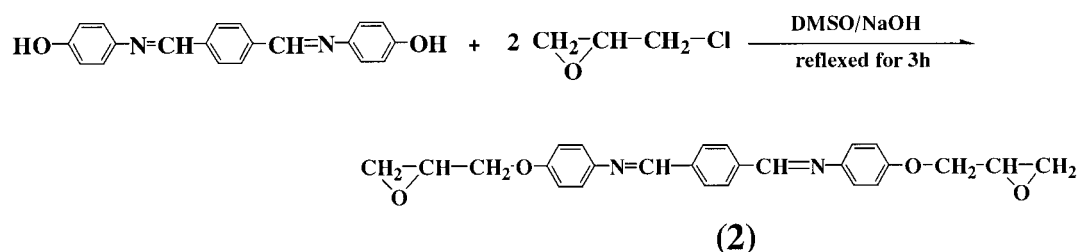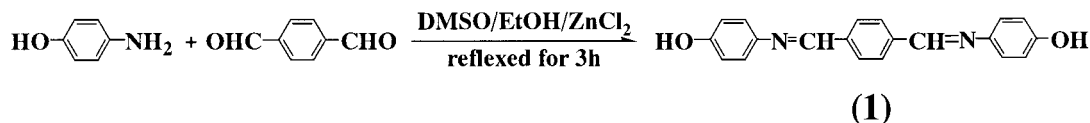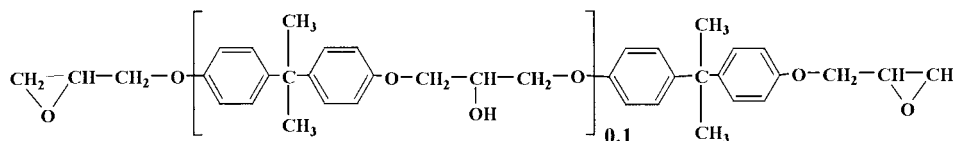
**Scheme 1** Synthetic pathway for DGETA.

crystalline epoxy resin. Moreover, the effects of the aliphatic chain length introduced in the backbone moiety on the bonding properties of the epoxy resin were also studied.
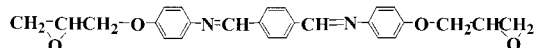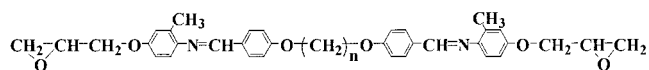
## EXPERIMENTAL

### Materials

The epoxy resins used were as follows:

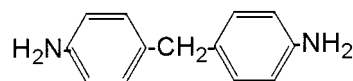

- The diglycidyl ether of terephthalylidene-bis(*p*-aminophenol) (DGETA; MW = 428).



- The twin mesogenic epoxy resins: diglycidyl ether of α,ω-bis(3-methylaminophenyloxy)alkane (*n* = 3; DGE3MA; *n* = 4; DGE4MA, *n* = 5; DGE5MA, *n* = 6; DGE6MA).



- 4,4′-Diaminodiphenylmethane (DDM, Kanto Chemical Co., Tokyo, Japan) was used as an aromatic amine curing agent.



### Synthesis of liquid-crystalline epoxy resins

The synthesis pathway for DGETA is shown in Scheme 1.

- The diglycidyl ether of bisphenol-A (DGEBA, Japan Epoxy Resin Co., Tokyo, Japan), the mesogenic epoxy resin.

Terephthalylidene-bis(*p*-aminophenol) (**1**) was synthesized from terephthalaldehyde and *p*-aminophenol in a solvent mixture of dimethylsulfoxide (DMSO) and ethanol using zinc chloride as a catalyst. 1-Chloro-2,3-epoxypropane reacted with the terephthalylidene-bis(*p*-aminophenol) in DMSO using tetra-*n*-butylammonium chloride as a catalyst to form the glycidyl ether of terephthalylidene-bis(*p*-aminohenol) (**2**). The structure of the synthesized compounds was confirmed by ^{1}H-NMR and FTIR spectroscopy.

^{1}H-NMR (CDCl$_3$): 2.77–2.93 ppm (m, 4H, epoxy CH$_2$); 3.37 ppm (m, 2H, epoxy CH); 3.97–4.27 ppm (d, 4H, CH$_2$—OPh); 6.95–6.99 ppm (d, 4H, aromatic); 7.98 ppm (s, 4H, aromatic); 8.53 ppm (s, 2H, CH=N). IR (KBr): 2900–2930 cm$^{-1}$ (CH$_2$), 1616 cm$^{-1}$ (CH=N), 1029 cm$^{-1}$ (ether), 914 cm$^{-1}$ (epoxy group).

The synthesis pathway for a twin mesogenic epoxy resin is shown in Scheme 2.

This synthesis consisted of three steps. The first step was a reaction for introducing the aliphatic chain into the backbone moiety. The intermediates (**3**) of the first step were synthesized from a dibromoalkane and hydroxybenzaldehyde in acetone. The second step was a
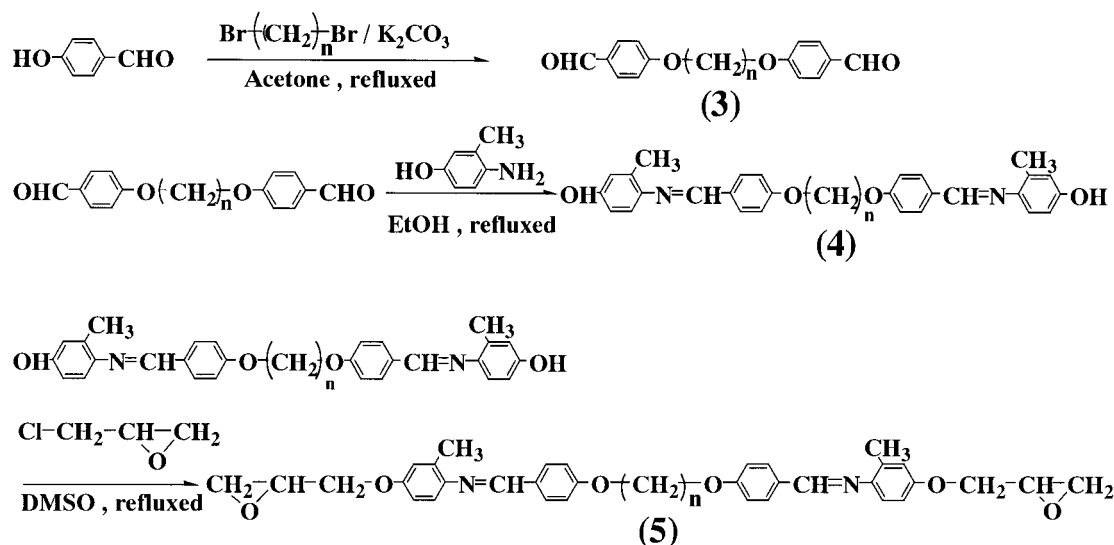
**Scheme 2** Synthetic pathway for twin mesogenic epoxy resins ($n$ = 3, 4, 5, and 6).

reaction for synthesizing the twin mesogenic diphenol (**4**). Finally, twin mesogenic epoxy resins (**5**) were synthesized by the epoxidation of the twin mesogenic diphenol (**4**) with 1-chloro-2,3-epoxypropane. The structure of the compounds synthesized was confirmed by $^1$H-NMR and FTIR spectroscopy.

DGE4MA: $^1$H-NMR (CDCl$_3$): 2.03 ppm (m, 4H, epoxy CH$_2$); 6.73–6.77 ppm (m, 4H, aromatic); 6.81–6.82 ppm (d, 2H, aromatic); 2.36 ppm (s, 6H, CH$_3$); 2.75–2.92 ppm (m, 4H, epoxy CH$_2$); 3.34–3.37 ppm (m, 2H, epoxy CH); 3.93–4.24 ppm (m, 4H, CH$_2$OPh); 4.11 ppm (s, 4H, CH$_2$); 6.89–6.99 ppm (m, 4H, aromatic); 7.82–7.85 (d, 4H, aromatic); 8.29 ppm (s, 2H, CH=N). IR (KBr): 2800–2930 cm$^{-1}$ (CH$_2$), 1621 cm$^{-1}$ (CH=N), 1029 cm$^{-1}$ (ether), 909 cm$^{-1}$ (epoxy group).

## Curing of epoxy resins

The terephthalylidene-type and twin mesogenic epoxy resins melted at 190 and 180°C, respectively. Stoichiometric amounts of the aromatic amine were then added as a curing agent. The former and latter resins were cured for 1 h at 190°C and 0.5 h at 180°C, respectively. The conversion of the epoxy group was followed during the curing process by FTIR spectroscopy.

## Measurement

The lap shear strength of joints was measured by using an Instron-type tensile machine (AGS-2000E Autograph, Shimazu Co., Kyoto, Japan) at a crosshead speed of 50 mm/min according to ASTM 1002-72. A steel plate was used as a substrate. The steel plate was degreased with acetone, abraded with polishing paper (#240), and then washed again with acetone. The morphology of the fracture surface after the lap shear test

was observed by using a scanning electron microscope (SEM: JSM-6500, JEOL, Tokyo, Japan) at an accelerating voltage of 15 kV. Before the examination, the surface was coated with a thin layer ($\sim$ 20 nm) of gold to improve conductivity and prevent charging.

Thermal analysis of the synthesized epoxy resins was carried out using DSC (DSC220C, Seiko Denshi Kougyou Co., Tokyo, Japan) at increasing temperature at a rate of 5°C/min in a nitrogen atmosphere. The sample weight was about 3 mg. Optical anisotropy of epoxy resin monomers was observed by using an optical microscope (BH-2, Olympus Co., Osaka, Japan) under cross-polarized light. The observation under the polarized microscope was carried out for the sample film mounted on a hot stage.

The internal stress of the cured epoxy resins was estimated from the deformation of the bimetal composed of the phosphor bronze plate (90 × 30 × 0.2 mm) and the cured epoxy resin coatings. As the coated resin was cured, the specimen was bent as a consequence of the difference between the thermal expansion coefficient of the phosphor bronze plate and the cured epoxy resin. The amount of displacement δ was measured with a laser displacement detector (LB-01, Keyence Co., Osaka, Japan). The internal stress $\sigma$ was calculated by the following equations:

$$\rho = \frac{l^2}{8\delta} + \frac{\delta}{2} \fallingdotseq \frac{l^2}{8\delta}$$

$$\sigma = \frac{E_1 h_1^3}{l2h_2} \frac{2}{\rho H}[1 + \frac{1}{3}(\frac{h_1}{H})^2]$$

where $E_1$ is Young's modulus of the phosphor bronze plate (1.22 × 10$^6$ kg/cm$^2$), $l2$ is the distance from a
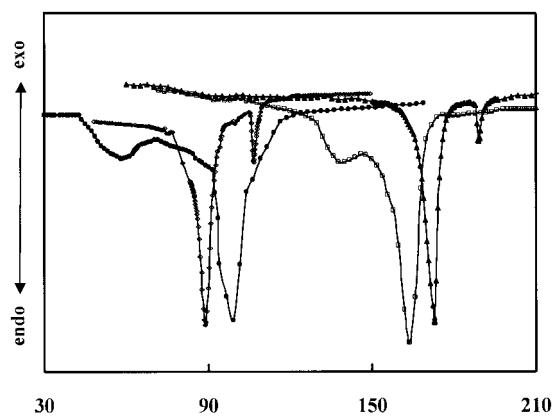
**Figure 1** DSC thermograms of twin mesogenic epoxy resins. Number of carbon atoms ($n$) of the aliphatic chains: (●) 3; (▲) 4; (◇) 5; (□) 6.



**Figure 2** Relationship between transition temperatures and length of aliphatic chain of twin mesogenic epoxy resins: (●) crystalline to nematic phase ($T_m$); (○) nematic to isotropic phase ($T_i$).

fulcrum to a measuring point; $h_1$ and $h_2$ are the thicknesses of a metal plate and coatings, respectively; $H$ is the total thickness ($h_1 + h_2$); $\delta$ is the displacement; $\rho$ is the radius of curvature; and $\sigma$ is the internal stress.

## RESULTS AND DISCUSSION

### Characterization of the liquid-crystalline epoxy monomers

DSC thermograms of four twin mesogenic epoxy resins containing different lengths of aliphatic chains between two mesogenic groups are shown in Figure 1. All curves show two endothermic peaks: the peak at the lower temperature exhibits the melting point ($T_m$) from crystalline to mesophase and the peak at the higher temperature exhibits the isotropic transition point ($T_i$) from the mesophase to the isotropic phase. These transition points, $T_m$ and $T_i$, of twin mesogenic epoxy resins are lower than those ($T_m$ = 195°C and $T_i$ = 215°C) of the terephthalylidene-type epoxy resin reported previously.[12] This result shows that the twin mesogenic epoxy resins synthesized in this work have a clear liquid-crystalline state and should melt at lower temperature. Thus, the twin mesogenic epoxy
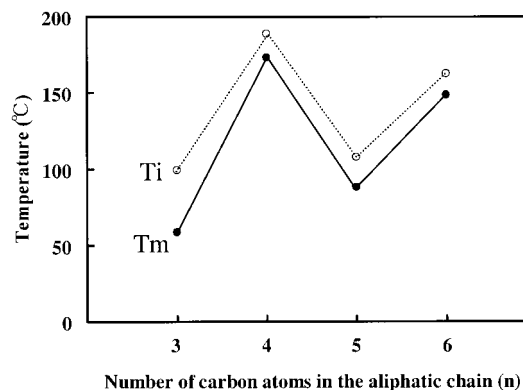
resins could be cured under milder conditions, compared with those of the mesogenic epoxy resin shown in previous reports.[5,12]

The relationship between the two transition temperatures, $T_m$ and $T_i$, and the number of carbon atoms of the aliphatic chains is shown in Figure 2. Both transition temperatures changed zigzag with an increase of the length of the aliphatic chains. This shows that the changes of the transition temperatures obey the rule of odd–even numbers,[15] which is well known in low molecular weight liquid crystals. In the epoxy resin that has an even number of carbon atoms between two mesogenic groups, the mesogenic groups have a *trans* configuration with each other and thus are closely packed in the crystalline state. This type of resin has higher phase-transition temperatures, $T_m$ and $T_i$, because of the close packing of the mesogenic groups. On the other hand, the epoxy resin that has an odd number of carbon atoms has lower transition temperatures because of the loose packing of the mesogenic groups. Thus, the twin mesogenic epoxy resins that have an odd number of the carbon atoms could be cured under milder conditions.

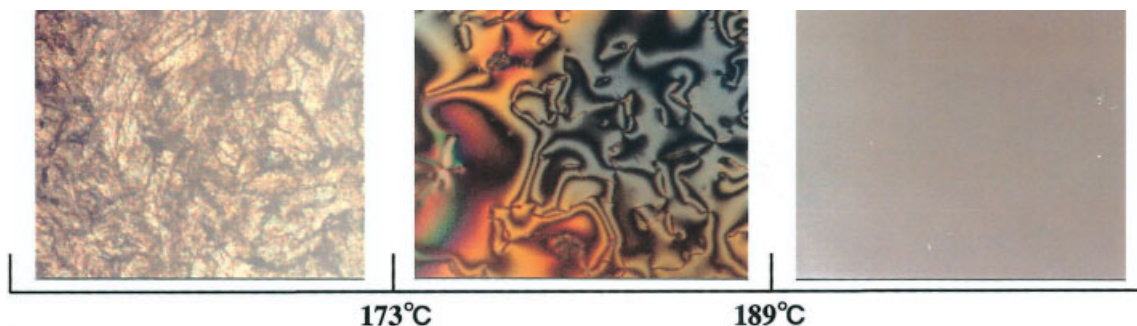The polarized micrographs of the twin mesogenic epoxy resin (DGE4MA) are shown in Figure 3. A



**Figure 3** Polarized micrographs of twin mesogenic epoxy resin ($n$ = 4, DGE4MA). Magnification ×100.
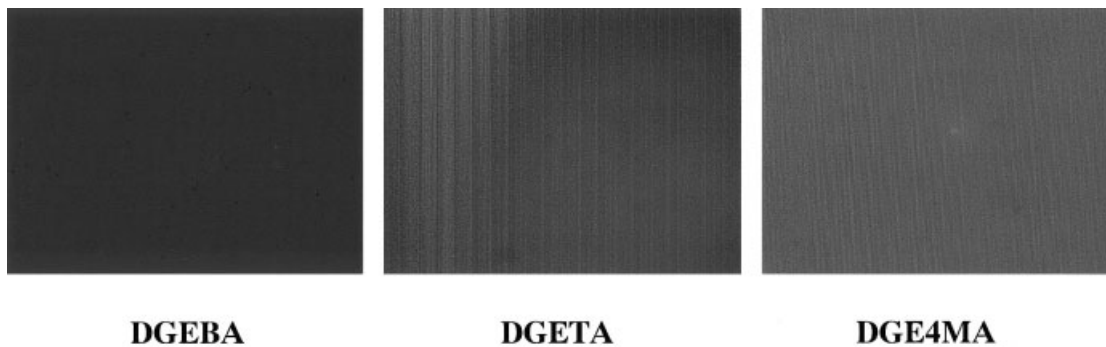
**Figure 4** Polarized micrographs of cured epoxy resin systems.

well-defined colored pattern, attributed to the orientation of the epoxy molecules, was observed in the temperature range < 189°C under cross-polarized light. The crystal phase was observed up to 173°C. The colored patterns, which are similar to a schlieren texture, were clearly observed at 173–189°C. This means that the DGE4MA is in the nematic liquid-crystalline state in the corresponding temperature regions. This colored pattern disappeared over 189°C. In this region, the epoxy resin is in the isotropic liquid state.

In addition, the same liquid-crystalline state was also observed in the other twin mesogenic epoxy resins that have different aliphatic chain lengths. Therefore, it is clear that all the twin mesogenic epoxy resins synthesized in this work could have the nematic liquid-crystalline state.

To study whether the twin mesogenic epoxy resin also has an ordered structure in the network structure, the DGE4MA/DDM cured system was observed under a polarized microscope. The polarized micrograph is shown in Figure 4 and was compared with that of the DGEBA/DDM system. Nothing could be seen under cross-polarized light in either of the DGEBA/DDM and DGE4MA/DDM systems, which means that both cured systems are isotropic materials and thus have no ordered structure. It is well known that the network chains of a cured epoxy resin containing a mesogenic group either has or does not have an ordered structure, which is analogous to that of a liquid crystal, when the epoxy resin was cured below or above some critical temperature, respectively.[16,17] If the mesogenic epoxy resin is cured at a high temperature, the resin monomer has high mobility during the progress of curing. In this study, the twin mesogenic epoxy resins were cured at a high temperature (190°C). At 190°C, the epoxy monomer should have very high mobility and thus would not be able to form an ordered structure in the cured resin. It must be stated that all the cured epoxy resins have no ordered structure analogous to that of a liquid crystal, even if the epoxy monomer itself shows a liquid-crystalline state in some temperature region.

## Dynamic mechanical properties of the cured twin mesogenic epoxy resins

Dynamic mechanical properties of the four twin mesogenic epoxy resins, which have different aliphatic chain lengths, are shown in Figure 5. The glass-transition temperature ($T_g$) of these cured systems appeared from 120 to 150°C and had lower values when the number ($n$) of carbon atoms, of the aliphatic chains, increased. This shows that the micro-Brownian motion of the network chains occurred more easily in the system that has longer aliphatic chains between two mesogenic groups. As shown in Figure 2, the phase-transition temperatures, $T_m$ and $T_i$, of the twin mesogenic epoxy resin monomers clearly obey the rule of odd–even numbers. However, this rule could not be applied to the glass-transition temperature of the cured resins. Because a curing reaction was carried out at high temperature, the network chains had no ordered structure in the cured resin. Thus, we consider that the glass-transition behavior of the cured twin mesogenic resin could not obey the rule of odd–even numbers.

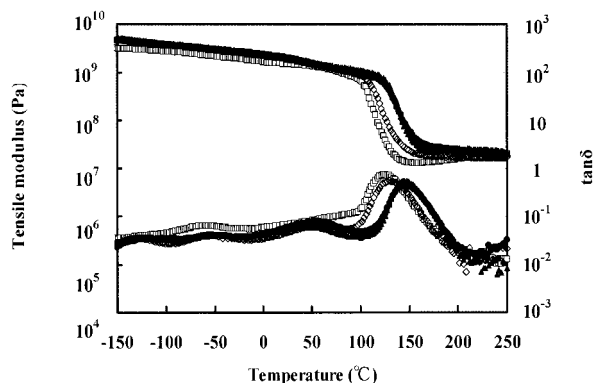Generally speaking, the glass transition of network polymers depends strongly on the chain mobility of



**Figure 5** Dynamic mechanical properties of cured twin mesogenic epoxy resins. Symbols are identical to those in Figure 1.
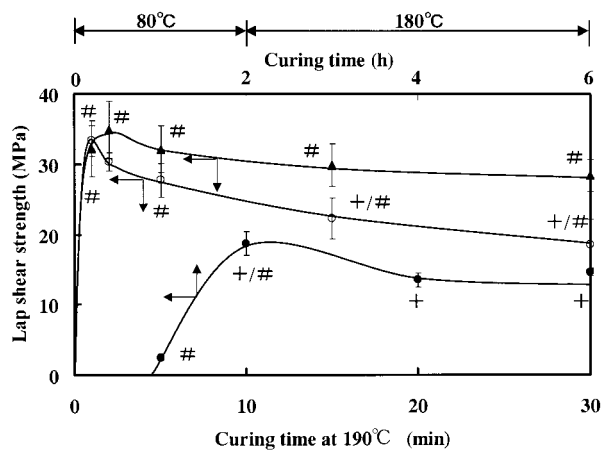
**Figure 6** Lap shear strength of epoxy resins cured with DDM. Epoxy resins: (●) DGEBA; (○) DGETA; (▲) DGE4MA. Fracture model: (+) contact failure; (#) cohesive failure.

the network chains. The $T_g$ values of the cured twin mesogenic epoxy resins studied in this investigation should also depend on the above factor because of their isotropic network structure. In the cured resin systems that contain short aliphatic chains, the conformation of two mesogenic groups should be restricted by the low mobility of the aliphatic chains and thus the cured resins must have a high glass-transition temperature. On the other hand, in the systems that have long aliphatic chains, the network chains should have high mobility and thus they must have a low glass-transition temperature.

## Bonding properties of liquid-crystalline epoxy resins

The lap shear strength in the curing process of the twin mesogenic (DGE4MA) and the mesogenic (DGETA) epoxy resins is shown in Figure 6, compared with that of the bisphenol-A–type epoxy resin (DGEBA). The bonding strengths of all the cured resin systems had maximum values at the initial stage of curing. In this stage, a cohesive fracture was observed

for all the cured resin systems. After that, the bonding strength decreased and then reached some constant value at the final stage of curing. In the final stage, the DGEBA system showed contact failure, the DGETA system showed a mixed fracture mode of contact and cohesive failures, and the DGE4MA system showed a cohesive fracture. As discussed in the previous report,[12] these results mean that, although the bonding strength should increase with curing, internal stress occurs with an increase in the curing shrinkage of the epoxy resin so that the bonding strength decreases after the vitrification of the cured resin. The decrement of the bonding strength in the twin mesogenic epoxy resin (DGE4MA) is lower than that of the DGETA and DGEBA systems. This result might show that the low internal stress of the DGE4MA system is lower than that of the other epoxy resin systems; thus the introduction of the flexible aliphatic chain into the backbone portion of the epoxy resin is effective for improving the bonding strength of the epoxy adhesive joints in the final curing stage.

To investigate the reason that the DGE4MA and DGETA systems have high bonding strength, the morphology of the fracture surfaces after the lap shear test was observed using a scanning electron microscope. The results of the SEM observations are shown in Figure 7. The fracture surface [Fig. 7(a)] of the DGEBA system showed a contact failure in the final stage of curing, and the adhesive layer showed no deformation in this resin system. This result shows that the fracture of the adhesive joints occurred at the interface between the epoxy resin and the substrate. On the other hand, the fracture surfaces of the DGETA system in the final stage of curing showed a mixed mode of cohesive and contact failures. Figure 7(b) shows the morphology of the cohesive failure region in this system. In the cohesive failure region of this system, the adhesive layer was largely elongated and then broken down. Generally, the cohesive fracture of the adhesive joints is caused when the deformation or fracture of the adhesive layer occurs more easily than the fracture at the interface. It was previously reported[12] that the
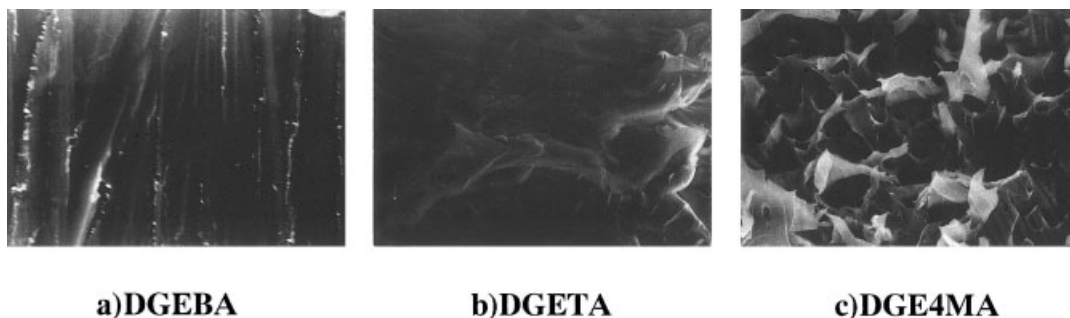


a)DGEBA      b)DGETA      c)DGE4MA

**Figure 7** SEM micrographs of fractured surfaces of epoxy adhesive joints: (a) DGEBA; (b) DGETA; (c) DGE4MA. Magnification ×500.
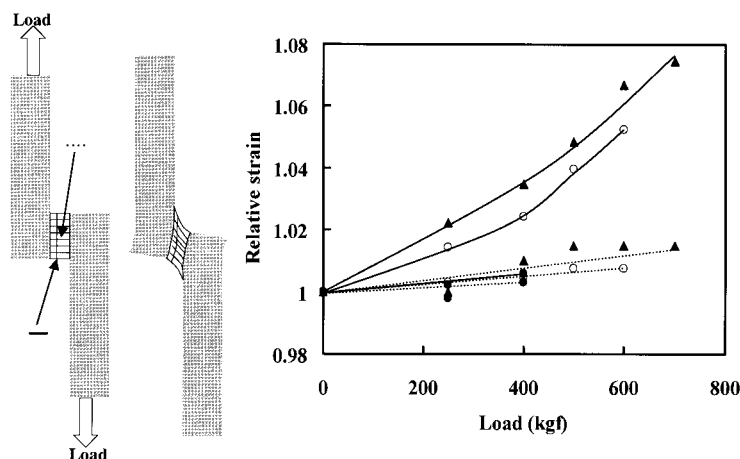
**Figure 8** Deformation of the adhesive layer in the lap shear test. Measuring point: (—) edge parts; (· · ·) center parts. Epoxy resins: (●) DGEBA; (○) DGETA; (▲) DGE4MA.

mesogenic epoxy resin DGETA cured with DDM has the same value of thermodynamic work of adhesion as that of the DGEBA/DDM system. Thus, this result should suggest that the high bonding strength of the DGETA was attributable to the large deformation of this system along the stress direction. The fracture surface [Fig. 7(c)] of the DGE4MA system also showed a cohesive failure with a large plastic deformation. This result shows that the plastic deformation of the DGE4MA cured system along the stress direction occurs more easily and the high bonding strength of the DGE4MA is a result of this plastic deformation.

To evaluate the deformational tendency of the adhesive layer during the lap shear test, the relative strains of the center and edge parts of the adhesive layer were observed for all the cured resin systems using a reading microscope. These results are shown in Figure 8. It is well known that the adhesive joint in the lap shear test was bent lightly with loading, as shown in this figure; thus the peeling component of the stress is generated in the adhesive joints. We measured the relative strain of the adhesive layer with the above deformation containing the bending of the joints. The relative strain of the center part of the adhesive layer with loading was very slight for all the cured resin systems because most of the load should be borne in the edge part of the adhesive joints. On the other hand, the relative strain of the edge part of the adhesive layer significantly increased with increasing load on the adhesive joints. Furthermore, the relative strain of the edge part of the DGE4MA system was greater than that of the DGEBA and DGETA systems. We reported in the previous study[12] that the deformation of the joints with the liquid-crystalline epoxy resin is greater than that of the bisphenol-A–type resin, and thus the load on the adhesive joint in the former resin system is able to disperse on the larger area of the bonding interface. This result showed that

the deformation of the adhesive joint bonded with the twin mesogenic epoxy (DGE4MA) system is considerably greater than that of the mesogenic epoxy (DGETA) system reported previously. This suggests that the network mobility of DGE4MA was improved to a greater extent, compared with that of the DGETA, because of the introduction of the flexible aliphatic chains into the epoxy network.

**Internal stresses of liquid-crystalline epoxy resins**

In the previous section, we described that the internal stress generated in the adhesive joint during the curing of the twin mesogenic epoxy resin might be lower than that of the other epoxy joint systems. To confirm the above deduction, the internal stress of the DGE4MA system in the cooling process after curing is shown in Figure 9, compared with that of the DGEBA and DGETA systems. The internal stress of all the cured resin systems was not generated in the rubbery region above $T_g$ of the cured resins. The internal stress was generated at the glass-transition region of the cured resins and then rapidly increased with a decrease of ambient temperature. In a previous report[18] we showed that the magnitude of the internal stress generated in the cooling process depends on the elastic modulus and expansion coefficient in the glassy region of the cured resin and also on the region of the cooling temperature. These results clearly show that the internal stress of the cured resins is mainly caused by cooling shrinkage in the glassy region below $T_g$, because of the high elastic modulus of the glassy epoxy resins. In addition, it is easily supposed that the internal stress is considerably relaxed in the transition region, given the high mechanical loss near the glass-transition region. Thus, we consider that the internal stress at the interface of the adhesive joint decreases
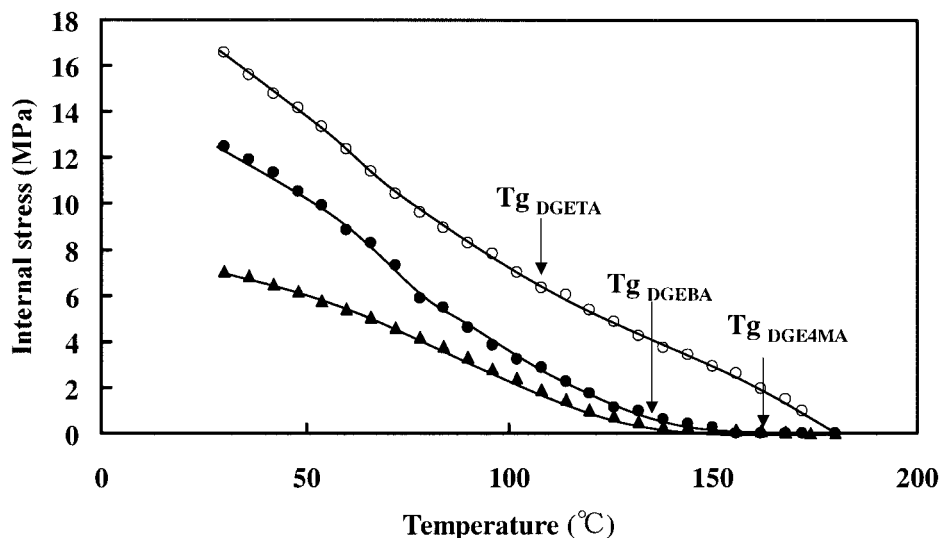
**Figure 9** Internal stress of epoxy resins occurring in the cooling process. Symbols are identical to those in Figure 4.

with an increase in the chain mobility of the epoxy networks.

The internal stress of the DGETA and DGE4MA systems was higher and lower, respectively, than that of the bisphenol-A–type epoxy resin (DGEBA). In a previous study[12] we showed that the liquid-crystalline epoxy resin (DGETA) has a large internal stress and thus the lap shear strength decreases with the progress of curing after the vitrification point. The introduction of the flexible aliphatic chain between two mesogenic groups clearly decreased the internal stress of the cured resin. Thus, it is reasonable to consider that the high lap shear strength in the final stage of curing of the DGE4MA system (Fig. 6) is attributed to the low internal stress of this system.

The expansion coefficient ($81.0 \times 10^{-6}\ °C^{-1}$) in the glassy region of the cured DGE4MA system had about the same value as those of the DGEBA ($75.5 \times 10^{-6}$ $°C^{-1}$) and DGETA ($74.5 \times 10^{-6}\ °C^{-1}$) systems. Thus, it is considered that the low internal stress of the twin mesogenic epoxy resin is not attributed to the modulus and expansion coefficient of the cured resin, but is a result of the low glass-transition temperature and the high mobility of the network chains in the cured DGE4MA system. That is, the internal stress of the DGE4MA system is lower than that of the other systems because of the narrow cooling width in the glassy region and the large stress relaxation in the transition region.

## Bonding properties of the twin mesogenic epoxy resins containing different aliphatic chains

The lap shear strength in the curing process of the four twin mesogenic epoxy resins that have different aliphatic chains is shown in Figure 10. The bonding

strengths of all the cured resin systems had maximum values near the vitrification point during the course of curing. After that, the bonding strength decreased slightly and then reached some constant value for each curing system. The lap shear strength after the vitrification point of all the twin mesogenic epoxy resins shows only a slight decrement, compared with that of the cured DGEBA and DGETA. In addition, cohesive fracture was observed for all the cured resin systems over the whole curing process. These results show that large internal stress does not occur, even after the vitrification in the curing process of the twin mesogenic epoxy resins. It is natural to consider that the low internal stress of the twin mesogenic epoxy systems is attributed to the large ability to relax stress by the flexible aliphatic chain introduced in the epoxy networks.
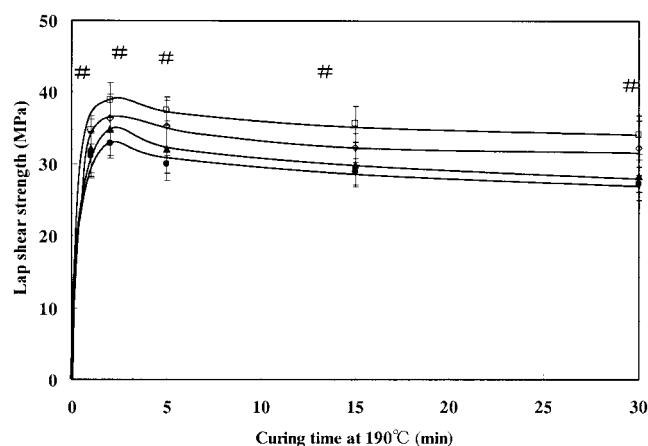


**Figure 10** Lap shear strength of cured twin mesogenic epoxy resins. Symbols are identical to those in Figure 1. Fracture mode: (#) cohesive fracture.
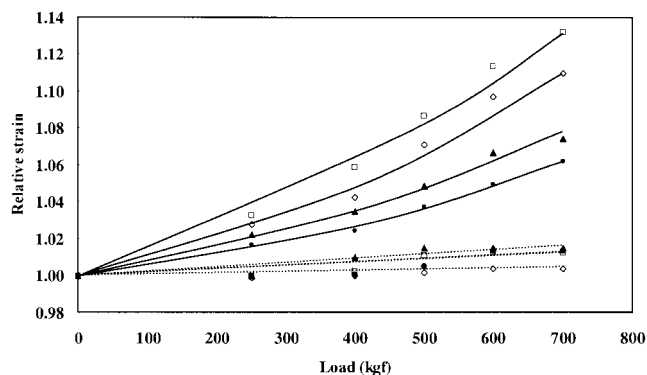
**Figure 11** Deformation of the adhesive layer in the lap shear test. Measuring point: (—) edge parts; (···) center parts. Symbols are identical to those in Figure 1.

The lap shear strength of the cured twin mesogenic epoxy resins increased with an increase in the length of the aliphatic chains. The cured system (DGE6MA) with the longest aliphatic chain showed the highest value of all the twin mesogenic epoxy resins. From this result, we conclude that the introduction of the flexible aliphatic chains between two mesogenic groups is effective for improving the bonding properties of the liquid-crystalline epoxy resins.

The relative strain of the adhesive layer at the center and edge parts of the adhesive joints was measured for all the twin mesogenic epoxy resin systems. These results are shown in Figure 11. Although the deformation of the center part of the adhesive layer with loading was very small for all the cured resin systems, the deformation of the edge part of the adhesive layer significantly increased with increasing load on the adhesive joints. This phenomenon displayed the same tendency as the deformation of the adhesive joints (Fig. 8) bonded with the DGEBA and DGETA. This result also shows that most of the load should be borne by the edge part of the adhesive joints. The relative strain of the edge part in the adhesive joints increased with an increase in the length of the aliphatic chains between two mesogenic groups. This means that the load on the joints is dispersed to a larger area and thus the lap shear strength of the joints showed higher strength with increasing network chain mobility, created by introducing longer aliphatic chains into the twin mesogenic epoxy resin.

## CONCLUSIONS

Twin mesogenic liquid-crystalline epoxy resins, containing an aliphatic chain between two mesogenic groups, were synthesized and their adhesive bonding strength was measured. Thus, we reached the following conclusions:

The introduction of the flexible aliphatic chains into the backbone moiety of the liquid-crystalline epoxy resin is effective for reducing the phase-transition temperatures, $T_m$ and $T_i$, of the mesogenic epoxy resin monomer. These transition temperatures obey the rule of odd–even numbers, which is well known for low molecular weight liquid crystals.

Twin mesogenic epoxy resin systems had a higher bonding strength than that of the commercial bisphenol-A and the mesogenic epoxy resins that were previously reported. The high bonding strength of the twin mesogenic epoxy resin system was attributed to the larger deformation of the resin along the stress direction. In addition, the small decrement of the lap shear strength after vitrification was observed in this resin system because of the low internal stress that occurred in the curing process.

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