Cycloaliphatic Epoxy Oligosiloxane-Derived Hybrid Materials for a High-Refractive Index LED Encapsulant

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ABSTRACT: Cycloaliphatic epoxy oligosiloxane resins with a high degree of condensation (>85%) were synthesized by a nonhydrolytic sol–gel reaction using 2-(3,4-epoxycyclohexyl)ethyltrimethoxysilane (ECTS), diphenylsilanediol (DPSD), and triphenylsilanol (TPS). Cycloaliphatic epoxy hybrimers with 2 mm thickness fabricated by thermal curing of cycloaliphatic epoxy oligosiloxane resins with a hardener and catalyst were optically transparent (~90%) with a high refractive index of up to 1.583. The fabricated hybrimers also show high thermal resistance having no yellowing during thermal aging at 120°C for 1008 h and a high decomposition temperature (>300°C). On the strengths of these characteristics, the hybrimers are expected to find application as LED encapsulants. © 2011 Wiley Periodicals, Inc. J Appl Polym Sci 122: 2478–2485, 2011

Key words: light-emitting diodes; oligomers; refractive index; transparency; UV–vis spectroscopy

INTRODUCTION

Conventional epoxy resins have been used to encapsulate light-emitting diodes (LEDs) for several decades, on the basis of strengths such as cost efficiency, good transparency, high refractive index, good adhesion, and good mechanical properties.[1,2] However, the heat budget is increased by the use of blue light with high intensity to realize a high-power white LED, leading to discoloration of epoxy encapsulants. Thus, due to low thermal stability against yellowing, epoxy resins are no longer used as the high-power white LED encapsulants. As a result, silicone resins, which exhibit higher thermal resistance, are recently being used instead.[3,4]

However, delamination between LED chips or wires and the silicone resins used as LED encapsulants in the module can occur due to the poor adhesive strength of the resins. Also, the conventionally used phenyl silicone resin has a low refractive index of ~ 1.53, thus resulting in limited light extraction efficiency of LEDs. Thus, in an effort to compensate for respective shortcomings of epoxy and silicone, epoxy silicone hybrid resins have been suggested for application as LED encapsulants, as they offer good adhesion and a higher refractive index than epoxy resins. However, in the cases of both silicone and epoxy hybrid silicone, the refractive index is limited below 1.55.[3–7]

Recently, we reported on the fabrication of cycloaliphatic epoxy-siloxane hybrid materials (hybrimers) by thermal curing of sol–gel synthesized cycloaliphatic epoxy oligosiloxane. Oligosiloxanes composed of both siloxane bonds and cycloaliphatic epoxy groups were synthesized by a simple nonhydrolytic sol–gel reaction of ECTS and DPSD. The fabricated transparent hybrimers showed a higher refractive index of up to 1.55 and better thermal stability against yellowing than epoxy resins for use as the LED encapsulants. Also, the hybrimers are expected to offer better adhesion and gas barrier characteristics compared to the silicone resins.[8,9] Thus, hybrimers are potential candidates for encapsulants in high-performance LEDs.

In this study, cycloaliphatic epoxy oligosiloxane resins containing a large amount of phenyl groups to increase the refractive index were synthesized by a nonhydrolytic sol–gel reaction of 2-(3,4-epoxycyclohexyl)ethyltrimethoxysilane (ECTS), diphenylsilanediol (DPSD), and triphenylsilanol (TPS). The synthesized cycloaliphatic epoxy oligosiloxane resins had high degree of condensation (DOC) and viscosity. *p*-Xylene was added as a solvent to improve processability of highly viscous cycloaliphatic epoxy oligosiloxane resins. The synthesized oligosiloxanes with a solvent were thermally cured with a hardener and catalyst to fabricate hybrimers
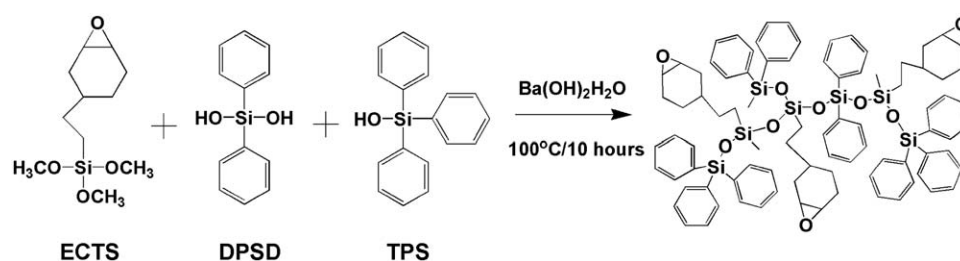
Scheme 1 Synthesis of cycloaliphatic epoxy oligosiloxane resins using TPS.

with a crosslinked network structure. The finally fabricated hybrimers show a high refractive index of over 1.58 at 632.8 nm and comparable thermal and mechanical properties and are thus feasible for use as an encapsulant for high-power white LEDs.

EXPERIMENTAL

Synthesis of cycloaliphatic epoxy oligosiloxane resins

Cycloaliphatic epoxy oligosiloxane resins were synthesized by a nonhydrolytic sol–gel reaction with ECTS (Gelest), DPSD (Gelest), and TPS (TCI) as precursors. Among these precursors, TPS was used to increase the refractive indices of the oligosiloxane resins. Barium hydroxide monohydrate ($Ba(OH)_2 \cdot H_2O$, 98%, Aldrich) was used as a catalyst to promote the nonhydrolytic sol–gel reaction between the precursors. The contents of $Ba(OH)_2 \cdot H_2O$ were 0.1 mol % of total silane. $Ba(OH)_2 \cdot H_2O$ was mixed in ECTS, and, alternately, DPSD and TPS were added several times at 100°C under N_2 purging for 2 h to obtain homogeneous solutions. After completing the addition of DPSD and TPS, the solutions were kept under the same conditions during a period of 10 h for additional reaction. The reaction scheme is shown in Scheme 1. The solutions were then cooled to room temperature and filtered using a 0.45-μm diameter Teflon filter to remove $Ba(OH)_2 \cdot H_2O$. We ultimately obtained cycloaliphatic epoxy oligosiloxane resins in the form of a clear solution. The formulations and notations of the synthesized oligosiloxane resins are listed in Table I.

Characterization of cycloaliphatic epoxy oligosiloxane resins

The formation of siloxane bonds and the cleavage of cycloaliphatic epoxy rings in the synthesized cycloaliphatic epoxy oligosiloxane resins were examined using ^{29}Si and ^1H nuclear magnetic resonance (NMR) spectrometers (600 MHz, DMX600, Bruker), respectively. The samples for the ^{29}Si and ^1H-NMR measurement were prepared with 30 vol % resins in chloroform-d_6 added chromium (III) acetylacetonate with a concentration of 30 mg L^{-1} as a relaxation agent of silicon. The viscosity of synthesized cycloaliphatic epoxy oligosiloxane resins was measured using rotational plate-and-plate rheometer (Rheostress 600, HAAKE). To measure viscosity of synthesized cycloaliphatic epoxy oligosiloxane resins, the samples were heated from 30 to 100°C with a 5°C/min heating rate. A cyclic shear stress of 1000 Pa was applied to the samples at 1-Hz frequency under a constant stress mode. Viscosity at 40°C was used from the measured viscosity-temperature curve. For a structural analysis of cycloaliphatic epoxy oligosiloxanes in the synthesized resins, FT-Raman spectra measurement of the resins was performed with a Bruker RFS 100 Raman-spectrometer equipped with a Nd:YAG laser (1064 nm) as an excitation source and a liquid-nitrogen-cooled, high-sensitivity Ge-detector. The spectra were measured with a laser power of 300 mW (100 scans per spectrum), a resolution of 2 cm^{-1}, and a spectral range between 50 and 3500 cm^{-1}. The distribution change of molecular species in the resins according to addition of TPS was analyzed using matrix-assisted laser desorption and ionization time-of-flight mass spectrometry (MALDI–TOF MS). MALDI–TOF MS spectra of the synthesized resins were obtained with a Voyager-DE STR 4700 proteomics analyzer equipped with a nitrogen laser using a wavelength of 337 nm and a pulse width of 3 ns of PerSeptive Biosystems. 2,5-Dihydroxybenzoic acid (Aldrich) and acetone were added as the matrix and solvent in the resins, respectively, for preparation of the measurement samples.

TABLE I
Formulations and Notations of Cycloaliphatic Epoxy Oligosiloxane Resins

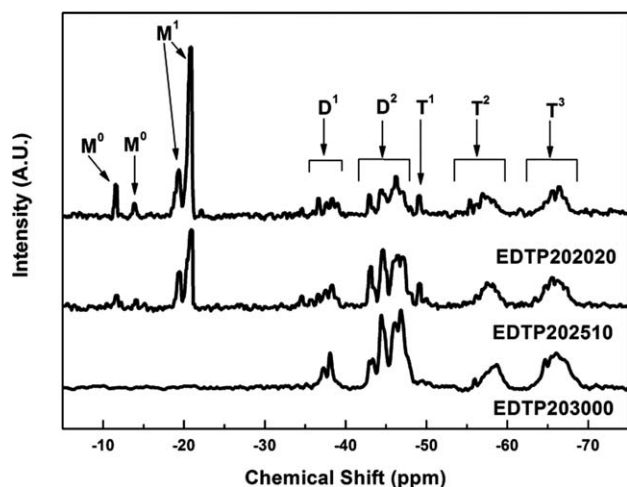| Resins	ECTS	DPSD	TPS	$Ba(OH)_2 \cdot H_2O$
EDTP203000	40.0 mol % (0.020 mol)	60.0 mol % (0.030 mol)	0.00 mol % (0.000 mol)	0.1 mol %
EDTP202510	36.3 mol % (0.020 mol)	45.4 mol % (0.025 mol)	18.2 mol % (0.010 mol)	0.1 mol %
EDTP202020	33.3 mol % (0.020 mol)	33.3 mol % (0.020 mol)	33.3 mol % (0.020 mol)	0.1 mol %

Figure 1 ^{29}Si-NMR spectra of cycloaliphatic oligosiloxane resins.

Fabrication of cycloaliphatic epoxy hybrimers

Methylhexahydrophthalic anhydride (MHHPA, 97%, Aldrich) and tetrabutylphosphonium methanesulfonate (TBPM, Fluka) were added as a hardener and a catalyst, respectively, to the synthesized oligosiloxane resins. The equivalent molar ratio of cycloaliphatic epoxy oligosiloxane resins and MHHPA was 1.0 : 0.9. The amount of TBPM was fixed at 0.5 mol % in MHHPA. *p*-Xylene (99%, Aldrich) was mixed as a solvent to reduce the viscosity of the synthesized cycloaliphatic epoxy oligosiloxane resins. The weight ratio between the synthesized resins and *p*-xylene was 1.0–0.1. The final resins were poured into a 2-mm thick mold fabricated with hexamethyldisilazane (Aldrich) treated glass. The mixed resins were kept at 50°C under a vacuum condition to remove bubbles and the solvent from the resins. The samples were thermally precured at 120°C for 2 h in an air condition. After the removal of the mold, they were thermally cured at 175°C for 12 h in a vacuum condition. Cycloaliphatic epoxy polymer was also fabricated with 3,4-epoxycyclohexylmethyl 3,4-epoxycyclohexanecarboxylate (Aldrich) using typical epoxy encapsulants for LED applications to compare thermal resistance between cycloaliphatic epoxy hybrimers to cycloaliphatic epoxy polymer.[4] The equivalent molar ratio of 3,4-epoxycyclohexylmethyl 3,4-epoxycyclohexanecarboxylate and MHHPA was 1.0–0.9. TBPM was added with molar ratio of 0.5 mol % in MHHPA. *p*-Xylene was not used due to low viscosity of the mixed cycloaliphatic epoxy resin. After the removal of bubbles in the resin, the mixed cycloaliphatic epoxy resin was thermally cured under the same curing condition.

Characterization of cycloaliphatic epoxy hybrimers

Thermal resistances of the fabricated cycloaliphatic epoxy hybrimers and cycloaliphatic epoxy polymer

were tested with optical transmittance change before and after thermal aging at 120°C for 1008 h under an air condition.

Transmittance spectra of the cycloaliphatic epoxy hybrimers and cycloaliphatic epoxy polymer were measured with an ultraviolet–visible-near infrared (UV/VIS/NIR) spectrophotometer (Shimadzu, UV3101PC) in a wavelength range of 300–800 nm. A thermogravimetric analyzer (TGA, TA instrument) was used to confirm the thermal stability of the hybrimers under N_2 condition with a heating rate of 5°C min. Shore *D* hardness of the hybrimers was defined using a shore *D* Durometer (HPSD, Schmidt). Their refractive index was measured with a prism coupler (Metricon, 2010) at a wavelength of 632.8 nm.

RESULTS AND DISCUSSION

Formation and characterization of cycloaliphatic epoxy oligosiloxane resins

Formation of siloxane bonds with thermal resistance in the cycloaliphatic epoxy oligosiloxane resins was confirmed with ^{29}Si NMR measurement (Fig. 1). Also, we calculated the DOC of the cycloaliphatic epoxy oligosiloxane resins with the peak area of ^{29}Si-NMR. M^n, D^n, and T^n denote the notation of Si from TPS, DPSD, and ECTS, respectively. The superscript "*n*" is the number of siloxane bonds of the Si atoms. The chemical shifts of the siloxane bond states are shown in Table II.[10–19] As shown in the ^{29}Si spectra, we confirmed that M species increase, and D species decrease with increment of TPS and reduction of DPSD, respectively. D^0 species are completely removed regardless of the addition of TPS. However, T^1 species slightly increase with the increment of TPS, and some unreacted M^0 species in the resins with addition TPS remain after the nonhydrolytic sol–gel reaction among the three precursors. This is attributed to the restriction of the nonhydrolytic sol–gel reaction by increase of bulkier phenyl groups in the resins according to the addition of TPS.

TABLE II
Chemical Shifts of Siloxane Bond States

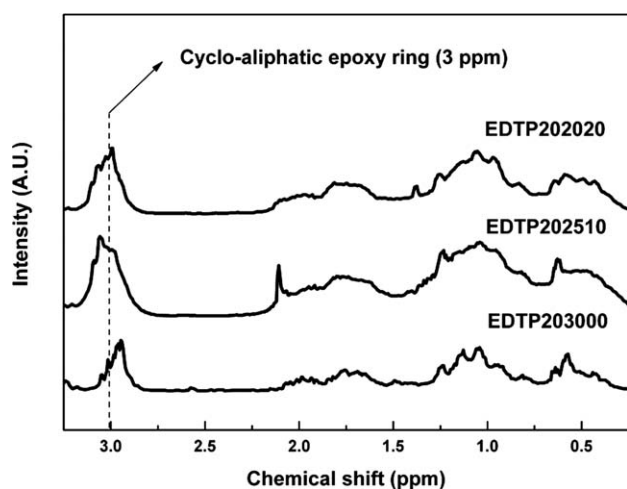| Species	Chemical shifts (ppm)
M^0 (triphenylmethoxysilane)	~-10.6
M^0 (triphenylsilanol)	~-12.4
M^1	~-17.5
D^0 (diphenyl dimethoxysilane)	~-29.0
D^0 (diphenylsilandiol)	~-34.0
D^1	-36.0 to -38.0
D^2	-42.0 to -47.0
T^1	-49.0 to -51.0
T^2	-57.0 to -61.0
T^3	-65.0 to -69.0

Figure 2 ^{1}H-NMR spectra of cycloaliphatic epoxy oligosiloxane resins.

The DOCs of the cycloaliphatic epoxy oligosiloxane resins were obtained with the following equation.[20–22]

$$
\text{DOC} = \frac{M^1 + D^1 + 2D^2 + T^1 + 2T^2 + 3T^3}{(M^0 + M^1) + 2(D^0 + D^1 + D^2) + 3(T^0 + T^1 + T^2 + T^3)} \times 100
$$

The DOCs of EDTP203000, EDTP202510, and EDTP202020 are 90%, 87%, and 86%, respectively. Despite the existence of M^0 species, the cycloaliphatic epoxy oligosiloxane resins show high DOCs of over 85%. Therefore, through analysis of the ^{29}Si-NMR spectra and the calculated DOCs, we confirmed that cycloaliphatic epoxy oligosiloxane resins were successfully synthesized.

The viscosity of EDTP203000, EDTP202510, and EDTP202020 is 51390.0, 639.0, and 588.9 Pa s at 40°C, respectively. The synthesized cycloaliphatic epoxy oligosiloxane resins show higher viscosity compared to previous cycloaliphatic epoxy oilgosiloxane resin (28.1 Pa s at 40°C) due to their high DOCs.[8] Therefore, it was difficult to handle these synthesized cycloaliphatic epoxy oligosiloxane resins relative to previous cycloaliphatic epoxy oilgosiloxane resin. We improved processability of highly viscous cycloaliphatic epoxy oligosiloxane resins by use of *p*-xylene as a solvent.

Ba(OH)$_2$•H$_2$O, which is used to promote the nonhydrolytic sol–gel reaction, is a basic catalyst. The cleavage of epoxy rings easily occurred both under a basic condition and by heat. It is necessary to confirm whether cycloaliphatic epoxy rings of the resins are open after the nonhydrolytic sol–gel reaction. ^{1}H-NMR spectra thus were measured to verify whether cycloaliphatic epoxy rings in the resins were cleaved (Fig. 2). In the ^{1}H-NMR spectra, chemical shift of the cycloaliphatic epoxy groups in the resins appears at 3 ppm. The chemical shift of 0.5 ppm originates from hydrogen atoms of carbon directly bonded at the silicon atom of ECTS. Through a comparison of the area of the chemical shifts, it is possible to verify whether cycloaliphatic epoxy groups in the resins are preserved.[21] As shown in the ^{1}H-NMR spectra presented in Figure 2, the area of the two chemical shift peaks is nearly the same regardless of their composition. Through the ^{1}H-NMR analysis, we confirmed that cycloaliphatic epoxy groups in the resins are preserved without ring opening under a basic condition induced by the addition of Ba(OH)$_2$•H$_2$O and at a temperature of 80°C.

Structural analysis of cycloaliphatic epoxy oligosiloxanes

Epoxy hybrid silicone and silicone resins used as encapsulants for white and high-power LEDs consist of polysiloxanes having a linear structure.[3–7] We measured FT-Raman spectra of the cycloaliphatic epoxy oligosiloxane resins to define their structure and access the differences between cycloaliphatic epoxy oligosiloxanes and polysiloxanes (Fig. 3). It has previously been reported that a three-membered ring peak of siloxanes exists at 606 cm^{-1}, and multimembered ring peaks of siloxanes were observed at a wavenumber under the three membered ring peak.[23,24] As shown in Figure 3, the three-membered rings of the cycloaliphatic epoxy oligosiloxanes are not shown at 606 cm^{-1} or at a wavenumber under the three-membered ring regardless of their compositions. Based on the FT-Raman results, we confirmed that the cycloaliphatic epoxy oligosiloxanes in the resins have linear structures, and their structure is the same as that of polysiloxanes consisting of epoxy hybrid silicone and silicone resins.
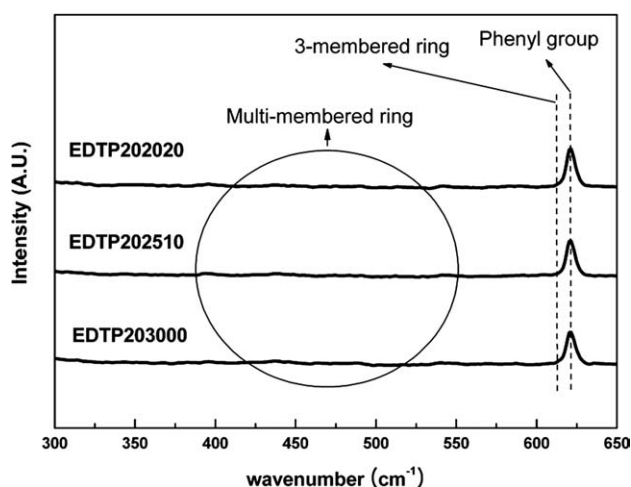

Figure 3 FT-Raman spectra of cycloaliphatic epoxy oligosiloxane resins.
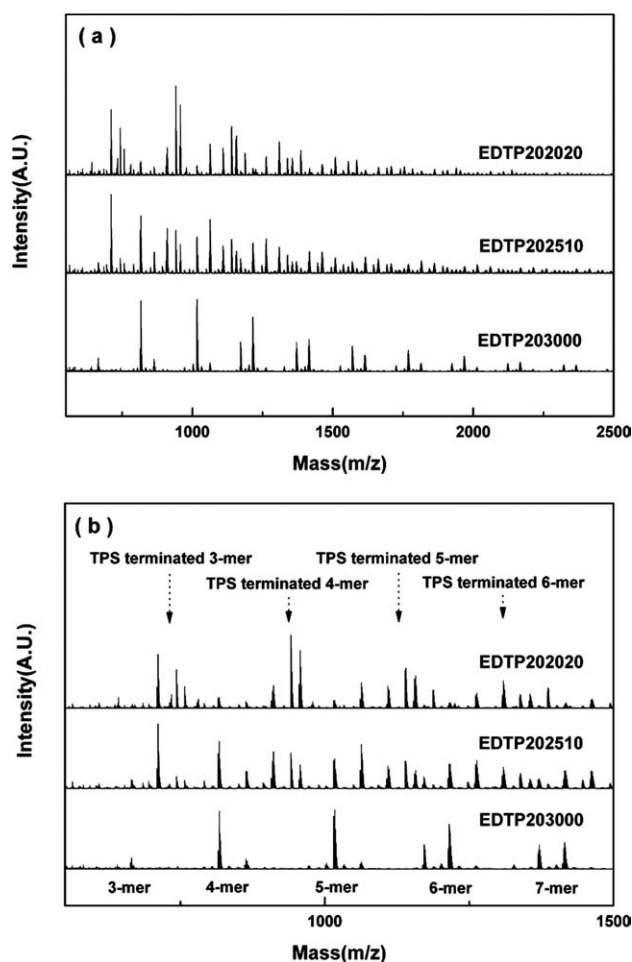
Figure 4 (a) MALDI-TOF mass spectra; (b) detailed spectra of cycloaliphatic epoxy oligosiloxane resins.

To confirm the molecular distribution of cycloaliphatic epoxy oligosiloxanes in the resins and support the FT-Raman results, we examined MALDI–TOF mass spectra of the cycloaliphatic epoxy oligosiloxane resins. TPS is terminated at the end of the cycloaliphatic epoxy oligosiloxanes, because TPS has one hydroxyl group. Therefore, it is possible to restrict the nonhydrolytic sol–gel reaction between ECTS and DPSD if TPS is preferentially terminated following the formation of the cycloaliphatic epoxy oligosiloxanes. However, as shown in the full MALDI–TOF mass spectra of Figure 4(a), the

molecular distribution of all cycloaliphatic epoxy oligosiloxane resins is under 2500 m/z regardless of the addition of TPS. In the detailed MALDI–TOF mass spectra of Figure 4(b), new peaks are observed between the peaks of the cycloaliphatic epoxy oligosiloxanes synthesized without TPS. The intensity of the new peaks increases and that of the previous peaks decreases with increment of TPS. With these results, we verified that the nonhydrolytic sol–gel reaction between the three precursors is promoted well without restriction by TPS.

It is also possible to verify the structure of the cycloaliphatic epoxy oligosiloxanes with their MALDI–TOF mass spectra. The practical molecular weight of each peak originating from the cycloaliphatic epoxy oligosiloxanes is provided in Table III. The molecular weight, calculated under the assumption that the cycloaliphatic epoxy oligosiloxane have a linear structure, is also summarized in Table III. Through comparison of the practical molecular weight with the calculated molecular weight, the FT-Raman spectra are supported. As shown in Table III, the peak positions of the practical molecular weight are consistent with those of the calculated molecular weight regardless of whether TPS is terminated at the cycloaliphatic epoxy oligosiloxane. Through comparison of the two peaks, we confirmed that the cycloaliphatic epoxy oligosiloxanes in the resins have a linear structure.

Optical and mechanical properties of cycloaliphatic epoxy hybrimers

The refractive index of a LED encapsulant is an important factor to realize high-luminescence efficiency in white and high power LEDs through enhanced light extraction efficiency. As shown in Figure 5, the refractive index of cycloaliphatic epoxy hybrimers increases with the increment of TPS. The cycloaliphatic epoxy hybrimers have a high-refractive index of over 1.56 without reference to their compositions due to the presence of many phenyl groups.[8] In particular, the cycloaliphatic epoxy hybrimer with TPS of 33.3 mol % shows a high-refractive index of over 1.58 at 632.8 nm. This is due to the presence of more phenyl groups, which increase the refractive index, in the cycloaliphatic epoxy hybrimer with higher

TABLE III
Comparisons Between the Practical Molecular Weights from the MALDI–TOF Mass Spectra and Calculated Molecular Weights from Molecular Weights of ECTS, DPSD, and TPS

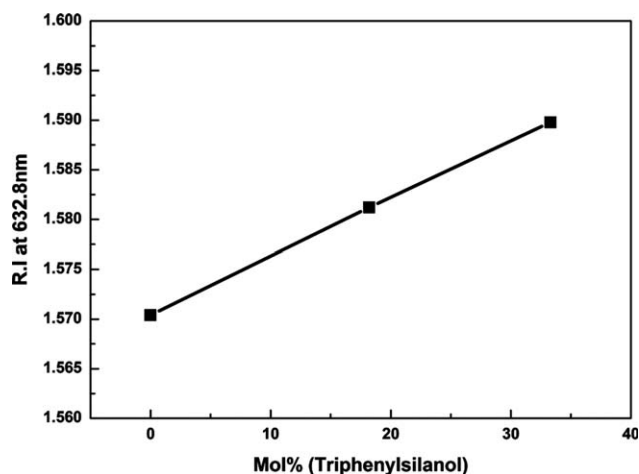|	Trimer	Tetramer	Pentamer	Hexamer	Heptamer	Octamer
No TPS terminated						
Calculated m/z	640–646	840–848	1038–1048	1236–1248	1434–1448	1632–1648
Practical m/z	663–669	815–868	971–1066	1169–1263	1325–1432	1524–1619
TPS terminated						
Calculated m/z	688–733	885–979	1084–1180	1282–1440	1480–1640	1678–1840
Practical m/z	685–792	907–960	1061–1161	1247–1358	1445–1588	1643–1757

Figure 5 Refractive index change of cycloaliphatic epoxy hybrimers as a function of TPS contents.

concentration of TPS. Because epoxy hybrid silicone and silicone resins, which are mainly composed of D species and T species, have limited amounts of phenyl groups, it is difficult to increase the refractive index beyond 1.55 at 632.8 nm.[4] However, we could realize a higher refractive index compared to that of epoxy hybrid silicone and silicone resins through the use of M species with phenyl groups.

LED encapsulants generally must have proper hardness to protect the compound semiconductor chip, which is used a source of blue light, as well as other components of the LED. The hardness of LED encapsulants is estimated by their Shore D hardness. Cycloaliphatic epoxy hybrimers fabricated with EDTP203000, EDTP202510, and EDTP202020 show a Shore D hardness of 86, 85, and 85, respectively. These values are higher relative to that of epoxy hybrid silicone resins (70–80) fabricated using a thermal cationic polymerization initiator and of silicone resins (60–75).[3,6] Through a comparison of cycloaliphatic epoxy hybrimers with epoxy hybrid silicone and silicone resins, we verified that the increased Shore D hardness of cycloaliphatic epoxy hybrimers originates from the use of cycloaliphatic epoxy groups and MHHPA as a hardener.

Thermal resistance and stability of cycloaliphatic epoxy hybrimers

Blue light with high intensity to make white and high-power LEDs generates heat as it passes through the LED encapsulant, and the created heat causes yellowness of the LED encapsulants. Because this yellowness could degrade the transmittance of blue light, which mainly has a wavelength of mainly 450 nm, the realization of white and high-power LEDs is challenging. Therefore, we evaluated the thermal resistance of cycloaliphatic epoxy hybrimers for use as encapsulants for white and high-power LEDs and

compared the thermal resistance of cycloaliphatic epoxy hybrimer to that of cycloaliphatic epoxy polymer. Cycloaliphatic epoxy hybrimers and cycloaliphatic epoxy polymers were thermally aged at
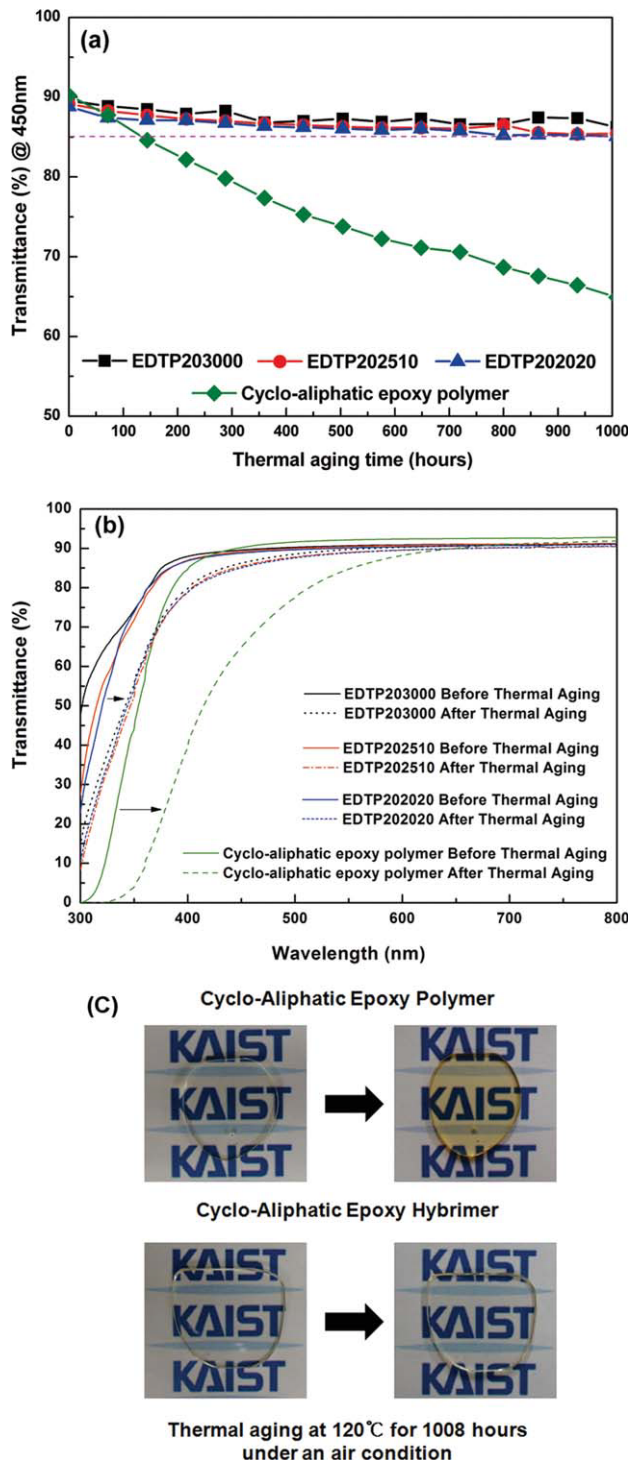

Figure 6 (a) Transparency at the wavelength of 450 nm according to thermal aging time at 120°C; (b) transmittance spectra before/after thermal aging at 120°C for 1008 h; (c) photographs before/after thermal aging at 120°C for 1008 h. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]
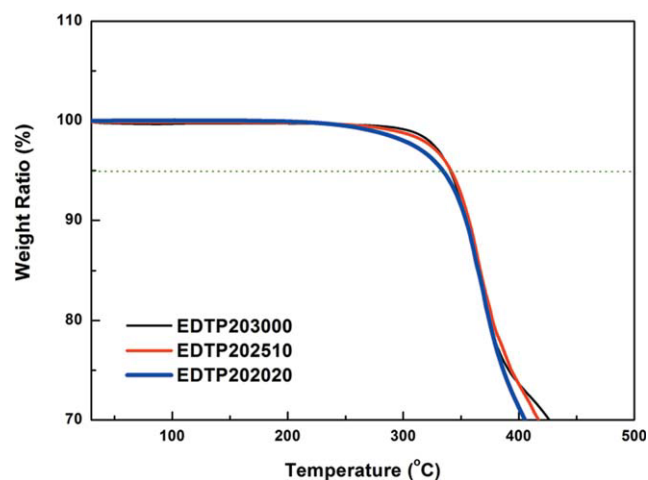
Figure 7 Thermal stability of cycloaliphatic epoxy hybrimers. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

120°C until 1008 h under an air atmosphere, and transmittances were measured every 72 h. As shown in Figure 6(a), the transmittances of the cycloaliphatic epoxy hybrimers at 450 nm were maintained over 85%, and the transmittance degradation was under 5% regardless of their compositions despite long thermal aging at 120°C under air. However, cycloaliphatic epoxy polymer showed transparency of under 65% after long thermal aging. Figure 6(b) represents the transmittance spectra of the cycloaliphatic epoxy hybrimers and cycloaliphatic epoxy polymer before/after thermal aging at 120°C for 1008 h to verify their capacity to withstand sustained thermal aging. Transmittances of the cycloaliphatic epoxy hybrimers at 450 nm are over 85%. Even though the transmittance of cycloaliphatic epoxy hybrimers is slightly degraded (<5%), the absolute values of decreased transmittance are outstanding compared to that of cycloaliphatic epoxy polymer generally used as a LED encapsulant. Figure 6(c) shows photographs of the cycloaliphatic epoxy hybrimers and cycloaliphatic epoxy polymer before/after thermal aging. While yellowness of cycloaliphatic epoxy polymer clearly appears in these photographs, discoloration of the cycloaliphatic epoxy hybrimer before/after thermal aging is not apparent in these photographs. This is attributed to the highly condensated siloxane network in the cycloaliphatic epoxy hybrimers.

Thermal stability is an important factor among the requirements for LED encapsulants, because heat produced in a LED device could generate outgassing of the encapsulants, and the performance of components in the device could be degraded by this outgassing. Figure 7 shows a TGA curve of cycloaliphatic epoxy hybrimers to assess their thermal stability. As shown in this TGA curve, the 5% weight loss temperature of cycloaliphatic epoxy hybrimer bulk is ∼ 340°C regardless of the composi-

tions. This high-thermal stability (>300°C) of cycloaliphatic epoxy hybrimer bulk is analogous to those of epoxy hybrid silicone resins and epoxy resins thermally cured by anhydride[5,8,25] and is suitable for use as a LED encapsulant.

CONCLUSIONS

Cycloaliphatic epoxy oligosiloxane resins with a high DOC (>85%) and a linear structure were successfully synthesized by a nonhydrolytic sol–gel reaction using ECTS, DPSD, and TPS. Cycloaliphatic epoxy hybrimers with 2 mm thickness were fabricated with the cycloaliphatic epoxy oligosiloxane resins, a hardener, and a catalyst. The fabricated cycloaliphatic epoxy hybrimers showed high transmittance (∼ 90%), thermal resistance against thermal aging at 120°C for 1008 h under air, high thermal stability (>300°C), and high-Shore D hardness (>80). The refractive index of the cycloaliphatic epoxy hybrimers increased with the addition of TPS due to the inclusion of a large amount of phenyl groups of TPS, compared to DPSD. The highest refractive index value achieved was over 1.58 at a wavelength of 632.8 nm. On the basis of these thermal, optical, and mechanical properties, cycloaliphatic epoxy hybrimers are suitable candidates for LED encapsulation.

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