

# Sol-gel synthesized linear oligosiloxane-based hybrid material for a thermally-resistant light emitting diode (LED) encapsulant

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The thermal resistance against discoloration of a high refractive index phenyl-siloxane hybrid material (hybrimer) was studied for LED encapsulation. The phenyl hybrimer was fabricated by a hydrosilylation reaction of linearly-structured vinyl oligosiloxane and hydrogen oligosiloxane resins using a platinum (Pt) catalyst. The linear oligosiloxane resins were synthesized by a non-hydrolytic sol-gel condensation reaction of di-functional silane precursors, containing vinyl, hydrogen, and phenyl groups. The oligosiloxane, having a higher degree of condensation and molecular size compared with the branched oligosiloxane, provided low shrinkage and an effective curing behavior. The phenyl hybrimer showed high optical transparency and a refractive index of up to 1.57. In particular, the amount of Pt catalyst was minimized to inhibit yellowing of the hybrimer under long-term aging at 180 °C. These results suggest that the linear oligosiloxane-derived phenyl hybrimer could be used as a high performance LED encapsulant.

## Introduction

Light-emitting diodes (LEDs) have increasingly been studied for application in displays and lighting fields, to fabricate environmentally friendly illumination devices with low energy consumption. The LED has replaced other sources of illumination due to its light extraction efficiency, long lifetime, and durability. Recently, the development of a LED encapsulant, which is used as a protective material for chips, wires, and phosphors against environmental effects, has become an important issue for the commercial use of LEDs. A long-term stable encapsulant with good optical characteristics is necessary, since the energy efficiency of the light emission is dependent on the transparency and refractive index of the encapsulant.<sup>1,2</sup>

The encapsulant has been produced by a thermal or photo polymerization of epoxy and silicon resins. Because of the heat generated during LED operation, siloxane-based encapsulants have recently been preferred as an alternative to epoxy-based encapsulants. The encapsulant appears to be effective in forming siloxane bonds which improve thermal resistance against yellowing.<sup>2-8</sup> In particular, a siloxane containing methyl groups shows high thermal stability and low refractive index, whereas a phenyl-siloxane has an improved refractive index due to the high polarizability of the phenyl groups.<sup>9</sup>

However, at high temperatures, the transparency of the phenyl-siloxane encapsulant can be reduced by yellowing, since the phenyl groups are susceptible to thermal oxidation and cleavage from the siloxane backbone.<sup>10</sup> Therefore, it is essential to design a LED encapsulant which is reliable at high temperatures and has great energy efficiency with a high refractive index.

Recently, we demonstrated that the newly developed sol-gel derived siloxane hybrid materials (hybrimers) were applicable as thermally stable LED encapsulants with a high refractive index.<sup>3-5</sup> The hybrimers offer the merits of both organic and inorganic materials such as optical transparency, refractive index tunability, good adhesion, and thermal and mechanical stability.<sup>11-14</sup> We reported on sol-gel synthesized vinyl oligosiloxane resin with a hydrosilane containing cross-linker and the fabrication of a silicon-based LED encapsulant. The residual hydroxyl (-OH) groups were reduced *via* a non-hydrolytic sol-gel reaction, to form highly condensed oligosiloxane with a branched structure. However, the resin showed bad processability due to its high viscosity, and thereby a solvent was needed to fabricate the encapsulant. There was a high level of shrinkage during curing, which was a critical problem for applying the resin to a LED packaging.<sup>4</sup>

In previous work, we introduced vinyl and hydrogen oligosiloxane resins, synthesized by sol-gel condensation of vinyltrimethoxysilane or methyldiethoxysilane with diphenylsilanediol (DPSD), used to fabricate the phenyl hybrimer for LED encapsulants.<sup>3</sup> We replaced the hydrosilane cross-linker with hydrogen oligosiloxane resin, to reduce volatility and to

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increase phenyl group content. We achieved a remarkable refractive index of up to 1.58, due to the large amount of phenyl groups attached to the siloxane backbone. Nevertheless, the phenyl groups are caged by the condensed siloxane network, inhibiting decomposition of the phenyl groups at high temperatures of up to 150 °C. However, an excess of catalyst was needed to promote the hydrosilylation reaction of the oligosiloxane resins, since they had a small molecular size and high steric hindrance derived from their branched structure. Though this bulky structure prevented the cleavage of the phenyl groups, the high amount of catalyst required became a crucial factor in the reduced thermal resistance at an elevated temperature over 150 °C.

Building on these works, we fabricated a novel hybrimer using a reduced amount of Pt catalyst for curing. To promote cross-linking of vinyl and hydrosilane groups, a linearly-structured vinyl oligosiloxane (LVO) resin was synthesized with an increased molecular size. The resin was derived from methylvinyl dimethoxysilane containing two methoxy groups to form linear linkages by reaction with DPSD, thereby showing low steric hindrance. The high degree of condensation was achieved with low levels of unreacted monomers, and functional groups. Due to these structural modifications, the hybrimer was successfully cured with minimized Pt catalyst content and low shrinkage. Most notably, an enhanced thermal resistance at 180 °C was achieved, which has not been reported to our knowledge for any commercial LED encapsulant having a high refractive index of up to 1.57. The novel phenyl hybrimer-based encapsulant can provide long-term stable optical clarity for high operating temperatures and high brightnesses of LEDs.

## Experimental

### Synthesis of the linear vinyl oligosiloxane resins

Methylvinyl dimethoxysilane (MVDMS, > 95%, TCI) and diphenylsilanediol (DPSD, 98%, Gelest) were used as precursors for the synthesis of linear vinyl oligosiloxane (LVO) resin. Vinyltrimethoxysilane (VTMS, 98%, Aldrich) and DPSD were used as precursors for the synthesis of vinyl oligosiloxane (VO) resin. Barium hydroxide monohydrate (BH, Aldrich) was added as a basic catalyst to promote the reaction (0.1 mol% of the total silane precursors). We synthesized the LVO resin using a sol-gel condensation of MVDMS and DPSD (1 : 1 molar ratio) at 60 °C for 12 h, then at 80 °C for 12 h whilst stirring and under N<sub>2</sub> gas purging. Also, the VO (1 : 1 molar ratio) resin, which was reported in our previous research, was synthesized.<sup>3</sup> An optimized composition of each of the silane precursors and degree of condensation (DOC) and viscosity of the synthesized resin are presented in Table 1. The hydrogen oligosiloxane (HO) resin was also synthesized for a hydrosilylation reaction with the LVO and VO resins, *via* a previously reported procedure.<sup>3</sup> The synthesized resins were filtered through a 0.45 μm Teflon filter and vacuum heated to remove the catalysts and the volatile component.

**Table 1** Composition and properties of the linear vinyl oligosiloxane and vinyl oligosiloxane resins

Resin	MVDMS/VTMS	DPSD	DOC	Viscosity
LVO	0.1 mol	0.1 mol	94.1%	4400 cP
VO	0.1 mol	0.1 mol	80.8%	4800 cP

### Fabrication of the phenyl hybrimer

The synthesized resins were thermally cured to fabricate the phenyl hybrimer. The LVO resin was mixed with the HO resin in a 1 : 1.2 weight ratio, and a 0.1 wt% (20 ppm) of Pt Karstedt's catalyst [Platinum(0)-1,3-divinyl-1,1,3,3-tetramethyl-disiloxane complex solution in xylene (~2% of Pt), Aldrich] was added. For the thermal hydrosilylation reaction between vinyl groups and hydrosilane groups, the mixed resins were cast into a glass mold (thickness = 2 mm), then heat cured at 170 °C for 4 h in air.

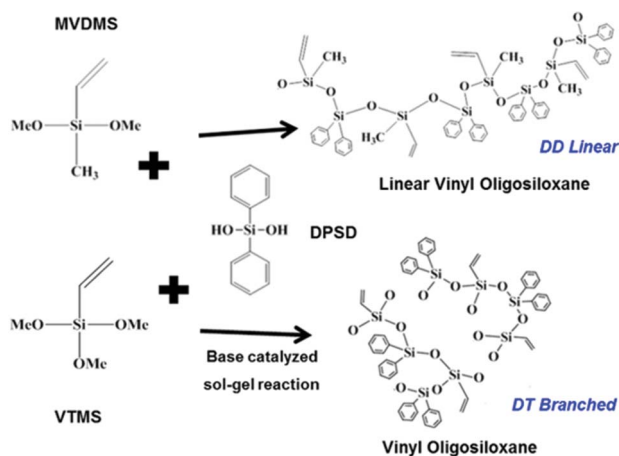
### Characterization of the phenyl hybrimer bulk and encapsulated LED

The <sup>29</sup>Si nuclear magnetic resonance (NMR) spectra of the oligosiloxane resins with chromium acetylacetonate (concentration of 30 mg L<sup>-1</sup>) in 30 vol% of chloroform-d were measured using a FT 600 MHz (Bruker Biospin, DMX600). Matrix-assisted laser desorption ionization time-of-flight (MALDI-TOF, Voyager-DE STR) was used to study the distribution of the molecular weights of the oligosiloxane resins with 2,5-dihydroxybenzoic acid as a matrix. Fourier transform-infrared (FT-IR) spectra (JASCO, FTIR 460 plus) and differential scanning calorimeter data (DSC, Netzsch DSC 200 F3 Maia) were measured in air to study the thermal hydrosilylation curing behavior of the mixed oligosiloxane resins with a Pt catalyst. A thermal gravimetric analysis (TGA, TA TGA 2050) was measured in a N<sub>2</sub> environment to measure weight loss during curing and thermal decomposition of the hybridizers. The optical transmittance and yellowness indices (YI, ASTM D1925) of the hybrimer bulks were measured using an ultraviolet-visible-near-infrared (UV/Vis/NIR) spectrophotometer (Shimadzu, UV-3101PC). The YI was calculated using a color analysis program. The refractive index of the hybrimer was obtained using a prism coupler (Metricon, 2010) at a wavelength of 632.8 nm. The shore hardness of the hybrimer bulk was measured using a shore durometer (Schmidt, HPSD). Water vapor permeability of the hybrimer (thickness = 2 mm) was characterized using a MOCON instrument (PERMATRAN-W MODEL 3/33). The photoluminescence (PL) intensity of the hybrimer encapsulated LED was measured using a PL spectrometer (DARSA PRO 5100 PL System, PSI trading Co., Ltd) and integrating sphere using a xenon lamp at room temperature.

## Results and discussion

### Formation of a linear vinyl oligosiloxane structure by a sol-gel reaction

A linear vinyl oligosiloxane (LVO) resin was synthesized through a sol-gel condensation reaction of di-functional



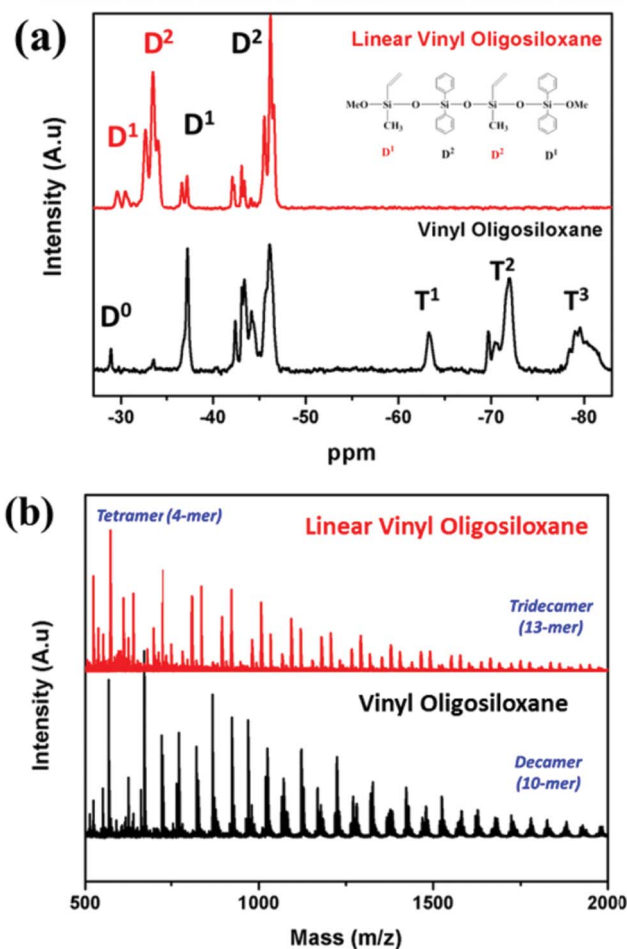
**Scheme 1** Synthesis of linear vinyl oligosiloxane (LVO) and vinyl oligosiloxane (VO) resins by sol-gel condensation reactions of methylvinyl dimethoxysilane (MVDMS)/vinyltrimethoxysilane (VTMS) and diphenylsilanediol (DPSD).

vinyl-methyl silane and phenyl silane precursors. As described in Scheme 1, methoxy ( $-\text{OCH}_3$ ) groups in the methylvinyl dimethoxysilane (MVDMS) reacted with silanol ( $\text{Si}-\text{OH}$ ) groups in the diphenylsilanediol (DPSD) through a nucleophilic attack by deprotonation of the hydroxyl ( $-\text{OH}$ ) groups, thereby forming covalently-bridged siloxane ( $\text{Si}-\text{O}-\text{Si}$ ) bonds.<sup>13–16</sup> Due to the cleavage of phenyl groups in acidic conditions, barium hydroxide (a basic catalyst) was used to promote the condensation reaction. Under a non-hydrolytic sol-gel process, silane monomers are directly condensed to form oligomers without water, which should be avoided to improve resin stability.<sup>17</sup>

The oligo-siloxane structure was confirmed by  $^{29}\text{Si}$  NMR and MALDI-TOF analysis as shown in Fig. 1. The chemical shift of the silicon atoms in the analyzed resin varies depending upon its bonding condition. The  $\text{D}^n$  and  $\text{T}^n$  represent Si atoms from silanes having 2 and 3 alkoxy groups, where  $n$  represents the number of siloxane bonds attached to a Si atom. The LVO resin contained only  $\text{D}^n$  species from MVDMS (red) and DPSD (black) precursors, resulting in the linear linkage of siloxane bonds. A high amount of  $\text{D}^2$  species ( $-32$  to  $-35$  and  $-42$  to  $-47$  ppm), which are fully condensed to have two siloxane bonds without any remaining methoxy or silanol groups, were clearly detected. From eqn (1), the calculated degree of condensation (DOC) of the LVO was 94.1%.<sup>18</sup>

$$\text{Degree of condensation} = \frac{\text{D}^1 + 2\text{D}^2}{2(\text{D}^0 + \text{D}^1 + \text{D}^2)} \times 100 \quad (1)$$

On the other hand, the vinyl oligosiloxane (VO) using T species silane showed a low DOC (80.8%), including a considerable amount of partially condensed  $\text{D}^1$ ,  $\text{T}^1$ , and  $\text{T}^2$  species. This is due to its branched (ladder-like) structure, and a high steric hindrance induced by the T species, inhibiting the additional formation of siloxane bonds by nucleophilic attack. We considered an inductive effect, which promotes the condensation reaction of silane monomers or oligomers. It is



**Fig. 1** (a)  $^{29}\text{Si}$  NMR spectra and (b) MALDI-TOF spectra of the linear vinyl oligosiloxane (LVO) and vinyl oligosiloxane (VO) resins showing the formation of highly condensed siloxane bonds with a molecular size distribution.

known that the partial charge of the methoxy group attached to the terminal location of the backbone is more negative than that of the methoxy group in an internal position.<sup>18</sup> Also, electron-withdrawing functional groups enhance the reaction rate in the base-catalyzed condensation, whereas in acid conditions cyclization occurs competing with the intermolecular condensation. The linear siloxane with high molecular weights and DOC can be formed due to the opening of rings and effective protonation of the methoxy groups produced in basic conditions.<sup>19,20</sup> Thus, there existed a few unreacted monomers ( $\text{D}^0$ ) and silanol groups ( $\text{Si}-\text{OH}$ ) in the LVO resin, which decreased resin stability by re-esterification or further condensation. These volatile contents in the resin create foams and shrinkage during curing, since the hydrosilane ( $\text{Si}-\text{H}$ ) groups react with hydroxyl ( $-\text{OH}$ ) groups to produce  $\text{H}_2$  gas.<sup>21</sup> Therefore, it is important to increase the DOC and minimize  $\text{D}^0$  and  $-\text{OH}$  groups in the resin, to fabricate a stable LED encapsulant.

As can be seen in Fig. 1b, we analyzed the linear siloxane structure and molecular weight ( $M_w$ ) using MALDI-TOF spectra. In order to increase the degree of cross-linking, it is

important to increase the size of the siloxane, thereby containing a large number of vinyl groups.<sup>22</sup> Compared with VO, the newly synthesized LVO consisted of oligomers with higher molecular size, from tetramer to tridecamer. Also, the empirical molecular weight of certain oligomers were well matched with the calculated  $M_w$  assuming the oligomer is a linear form. The probability of ring closure of linear siloxane is controlled by the presence of bulky phenyl substituents. Cyclosiloxanes are also susceptible to cleavage in basic conditions.<sup>20</sup> Therefore, we concluded that the LVO was successfully condensed to form a linear siloxane structure with high molecular size and large vinyl group content. The linear structure and high molecular size of the LVO can help an effective cross-linking due to its lowered steric hindrance and increased vinyl functionality. Even though the LVO resin had increased DOC, it still showed low viscosity due to its linear structure, which is appropriate for LED encapsulant fabrication.

### Curing of the linear vinyl oligosiloxane with hydrogen oligosiloxane resins

As mentioned above, the synthesized linear vinyl oligosiloxane resin had a small amount of remaining functional groups, but a large number of vinyl groups with low steric hindrance. We fabricated a phenyl hybrimer (PH) by a hydrosilylation reaction of LVO and hydrogen oligosiloxane (HO) resins (as described in Scheme 2). The hydrosilylation is a dominant curing system for LED encapsulants, since it is an additional cure showing low shrinkage and releases no by-products. A platinum (Pt) catalyzed thermal hydrosilylation reaction causes cross-linking of vinyl (C=C) and hydrosilane (Si-H) groups, linking  $\pi$ -bonds in the vinyl group and  $\sigma$ -bonds in the hydrosilane group through  $\pi$ - $\sigma$  rearrangement.<sup>23,24</sup>

The LVO resin was mixed with HO resin, then thermally cured with a Pt catalyst. The results discussed in previous studies using a VO resin indicated that the branched siloxane structure needed a high amount of catalyst for the hydrosilylation reaction. An excess of Pt catalyst in the cured material has an adverse effect on thermal resistance against discoloration due to its high levels of light absorption and the formation of Pt<sup>0</sup> colloids. Also, free radicals, formed by an oxygen and the remaining catalyst, are known to be associated with the thermal degradation of organic components.<sup>17</sup> In this study, we minimized the content of the catalyst to 0.1 wt% (20

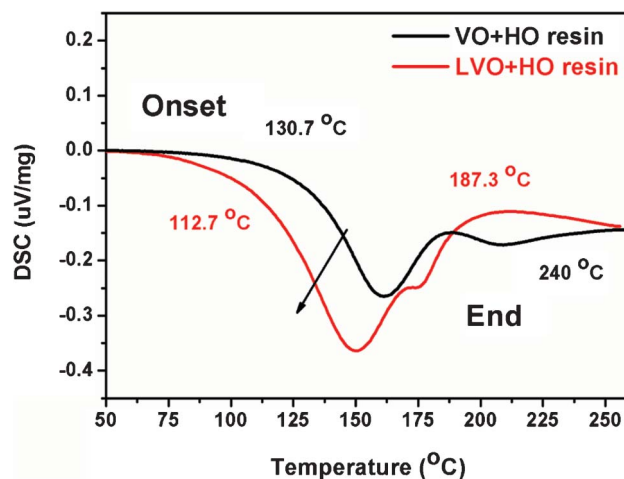


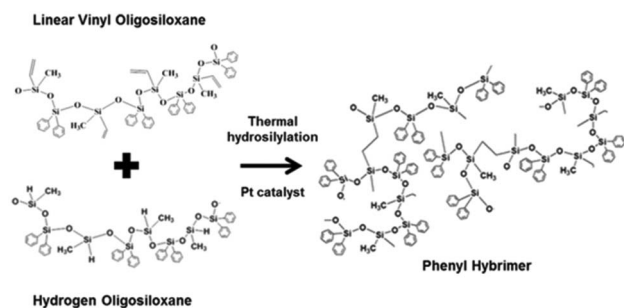
Fig. 2 DSC spectra of mixed resins of LVO or VO with HO and 20 ppm of Pt catalyst.

ppm) of the mixed LVO and HO resins, in order to improve the thermal resistance and decrease shrinkage of the hybrimer. The Pt catalyst content in LVO was a tenth of that of the previously fabricated phenyl hybrimer using VO and HO resins (200 ppm).

Fig. 2 shows the results obtained using DSC, which indicates the curing behavior of the LVO and VO resins with the same Pt catalyst content (20 ppm). The spectra shows an exothermic cross-linking of the resin by hydrosilylation. The onset curing temperature decreases from 131 °C to 113 °C by modifying the structure and size of the vinyl oligosiloxane. Also, the enthalpy change ( $\Delta H$ ) of the VO resin is low compared with that of the LVO resin, and the reaction continues to a temperature of 240 °C. Due to the steric consequences of internal vinyl groups in the VO structure, the cross-linking did not fully occur in the VO resin and the rate of the curing was slower than that of the LVO resin. The rate of the hydrosilylation is attenuated by the bulky structure of T type siloxane. Un-crosslinked vinyl groups or phenyl groups can be the source of the discoloration of the hybrimer during thermal aging.<sup>17</sup> Thus, we need a very high temperature and long curing time for the fabrication of a phenyl hybrimer using bulky VO resin with 20 ppm of Pt catalyst content.

The mixed LVO and HO resins containing 20 ppm of catalyst were cured to fabricate the phenyl hybrimer, which was confirmed by FT-IR analysis as shown in Fig. 3. The band at 916  $\text{cm}^{-1}$  is assigned to  $=\text{CH}_2$  bonds in the vinyl groups, and bands at 870–916  $\text{cm}^{-1}$  are assigned to  $-\text{O}-\text{Si}-\text{H}$  bonds in the HO resin.<sup>25</sup> After thermal curing at 170 °C, these bands completely disappeared, due to the cross-linking between the vinyl groups and hydrosilane groups in the resin. On the basis of these analyses, the LVO resin is appropriate for the fabrication of a LED encapsulant containing a small amount of catalyst.

We also measured weight loss (shrinkage) of the mixed resins of LVO and VO under curing conditions, as shown in Fig. 4. The shrinkage and bubbles during curing of the LED encapsulant originated from the volatile components in the



Scheme 2 Fabrication of a phenyl hybrimer (PH) using LVO and HO resins by a thermal hydrosilylation reaction.

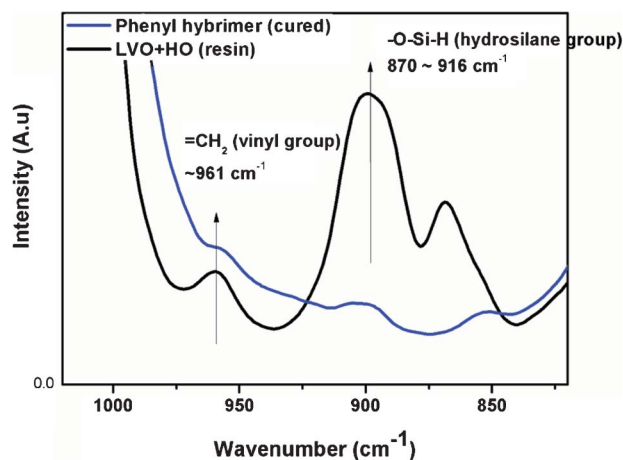


Fig. 3 FT-IR spectra before and after curing of the phenyl hybrimer fabricated with LVO and HO resins using a Pt catalyst.

resin, such as  $-OH$  groups, unreacted monomers, low molecular weight cyclic oligomers and solvent in the catalyst. Thus, we aimed to form highly condensed siloxane bonds with high molecular size and to decrease the catalyst content. Furthermore, we achieved complete cross-linking by introducing a linear structure, to minimize volatility during the curing process. When we used 20 ppm of catalyst, the LVO resin shows little weight loss (0.5%) from its initial state, while the VO resin had higher volatility (2.1% loss). The decreased weight loss was associated with reduced bubbles trapped inside the encapsulant and reduced shrinkage deformation of its initial shape after the curing process.

We studied the thermal decomposition stability of the fabricated PH bulk using TGA analysis, shown in Fig. 4. The PH fabricated using LVO has improved thermal stability showing a higher 5 wt% decomposition temperature (431.1 °C) compared with PH fabricated using VO (420.0 °C). The results showed us an increased cross-linking density of the hybrimer formed using LVO due to its effective curing behavior. Also, the decreased amount of unreacted components such as methoxy and silanol groups in the oligosiloxane delayed thermal decomposition. In order to cure the VO resin, we needed a higher temperature ( $>200$  °C) and a longer reaction time, which should be avoided due to the degradation of the LED performance. Therefore, we replaced the VO resin with LVO resin to fabricate a LED encapsulant containing a low amount of Pt catalyst for improved thermal resistance at a higher temperature, and reduced shrinkage during curing.

#### Thermal resistance of the phenyl hybrimer and long-term reliability of the PH-encapsulated LED

We prepared a highly cross-linked linear siloxane backbone in the phenyl hybrimer, fabricated by the curing of LVO and HO resins with a 20 ppm of Pt catalyst content. Then, a thermal resistance test of the PH bulk was performed at 180 °C for 1000 h with the commercial high refractive index silicone LED encapsulant (OE-6630, Dow Corning) as a reference. In a prior work, we reported excellent thermal resistance (at 150 °C) of

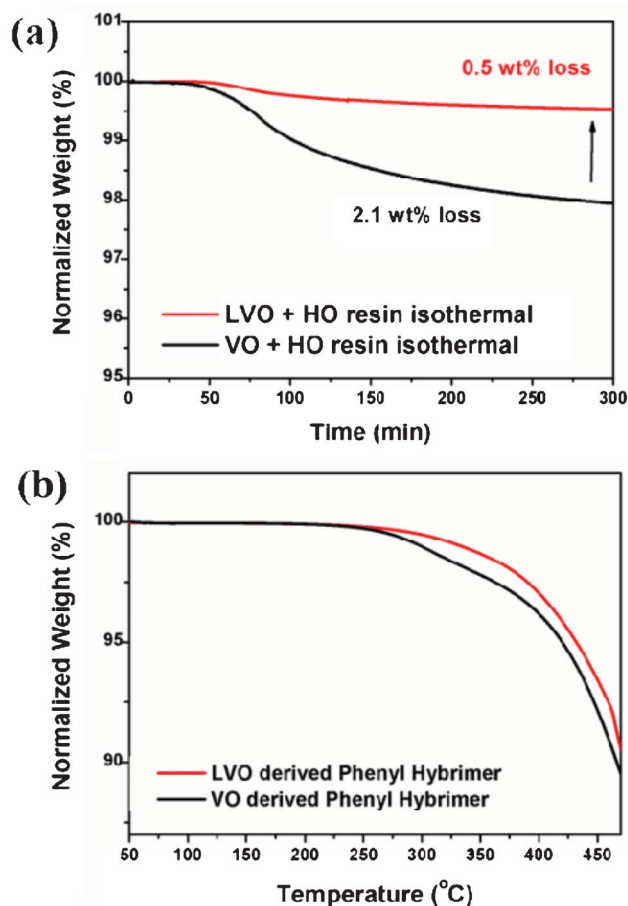


Fig. 4 (a) Isothermal TGA analysis of mixed resins of LVO or VO with HO, with a Pt catalyst at 170 °C for 4 h in air. (b) TGA analysis of the phenyl hybrimer bulk fabricated using LVO or VO resins.

the phenyl hybrimer fabricated by the curing of VO and HO resin.<sup>3</sup> However, a yellowing at an elevated temperature (180 °C) was observed due to its high catalyst content (200 ppm) and the presence of unreacted groups. Thus, we fabricated the PH using highly condensed LVO containing 20 ppm of catalyst to improve thermal resistance against discoloration.

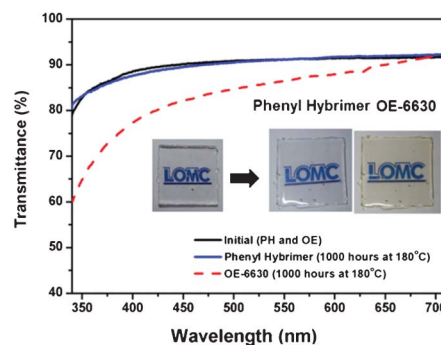


Fig. 5 Transmittance spectra and photo images of the phenyl hybrimer and OE-6630 (Dow Corning) bulk before/after thermal aging at 180 °C for 1000 h. The thickness of the bulk was 2 mm.

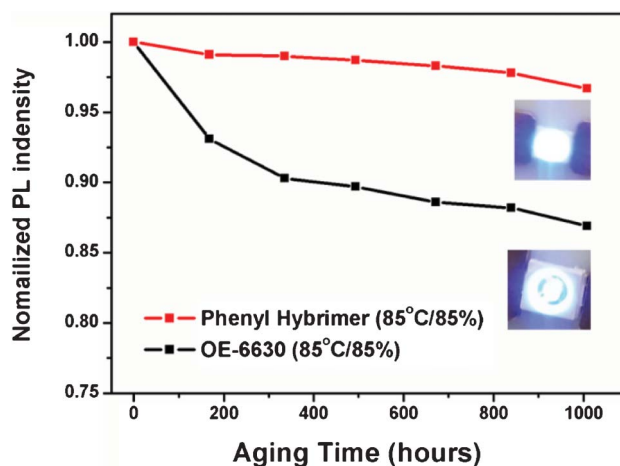
**Table 2** Characteristics of the newly fabricated phenyl hybrimer

Property	Unit	Value
Transmittance	% (at 450 nm)	89.7
Refractive index	n (at 632.8 nm)	1.571
5 wt% loss temperature	°C (5 °C min <sup>-1</sup> , N <sub>2</sub> )	431.9
Hardness	Shore D	60
Water vapor permeability	g m <sup>-2</sup> day <sup>-1</sup>	3.4

As shown in Fig. 5, the optical transmittance of the PH and OE-6630 was initially about 90% (at 450 nm) before thermal aging. The transmittance of the PH was almost maintained after 1000 h of thermal aging, while the OE-6630 showed a decrease in transmittance of 11%. The drop in optical transparency was due to the yellowing of the bulk, whereas the discoloration of the PH was hardly observed by the naked eye. A yellowness index (YI) based on ASTM, D1945 analysis, was also calculated to compare the degree of discoloration. The yellowness index change ( $\Delta YI$ ) of the hybrimer was 0.6, which was a much smaller value than that of the OE-6630 (8.9). As we determined, the highly cross-linked PH from highly condensed LVO resin, provided less formation of free radicals originating from monomers, low molecular weight oligomers, unreacted silanol or methoxy groups, and un-crosslinked organic groups. Due to the increased degree of condensation and cross-linking, the linear siloxane backbone also inhibited the cleavage of phenyl groups.<sup>26</sup> Our results provided evidence for the remarkable thermal resistance of the phenyl hybrimer at high temperatures up to 180 °C. As a consequence, the PH encapsulated LED, which was free of yellowing, had an advantageously long lifetime under high junction temperatures.

The various characteristics of the phenyl hybrimer, newly fabricated by curing LVO and HO resin with a Pt catalyst, are listed in Table 2. The high refractive index of the PH gives better LED light extraction efficiency. Due to the highly condensed Si-O bonds and phenyl groups in the cured hybrimer, we obtained a higher refractive index, up to 1.571, compared with that of OE-6630 (1.53). The hardness of the PH was tunable by varying the mixing ratio of the vinyl and hydrosilane groups to fabricate soft or hard encapsulants. To protect the LED from moisture, a lower water vapor permeability was also achieved compared with that of other linear siloxane materials such as polydimethylsiloxane.<sup>27,28</sup>

We fabricated PH- and OE-6630-encapsulated blue LEDs to verify reliability at high temperature (85 °C) and humidity (85% RH) conditions. While the LEDs were aged at 85 °C and 85% humidity, a photoluminescence (PL) intensity at 450 nm wavelength was measured for 1000 h. Fig. 6 shows a noticeable drop in PL intensity of the OE-6630-encapsulated LED. The PH-encapsulated LED maintained its initial intensity, suggesting that the PH encapsulant has good thermal resistance, adhesion, and barrier properties. There was no crack or delamination of the encapsulating hybrimer, showing excellent adhesion to the LED chip due to reduced shrinkage and bubbles during curing. On the basis of these analyses, we found that the high refractive index PH encapsulant could be



**Fig. 6** Normalized PL intensity change of the blue LED encapsulated by the phenyl hybrimer and a commercial high refractive index silicone encapsulant (OE-6630, Dow Corning), during aging at 85 °C and 85% relative humidity for 1000 h in air.

applied to a practical LED device due to its remarkable durability in harsh operating conditions.

## Conclusions

A novel phenyl-siloxane hybrid material was fabricated by a hydrosilylation reaction of linear oligosiloxane resins with a low Pt catalyst content. We modified the structure of the previously reported vinyl oligosiloxane, to synthesize highly condensed linear oligosiloxane with a large molecular size. We confirmed that the hybrimer showed remarkable thermal resistance against yellowing, compared with a commercial high refractive index silicone encapsulant. The hybrimer achieved not only a high refractive index but also a better thermal resistance through the structural modification of the oligosiloxanes. These results indicate that the sol-gel derived oligosiloxane-based hybrimer is a promising candidate material for high performance and reliable LED encapsulants.

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