

## Thermo-mechanical and Light Transmittance of Silica Diffusant Filled Epoxy Composites

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**Abstract:** Epoxy ternary blends (DCN) were prepared by mixing diglycidyl ether bisphenol A (DGEBA), cycloaliphatic epoxy, and novolac epoxy. The silica diffusants were prepared by the addition of spherical silica (SS) into epoxy blends. The thermal properties of the epoxy composites were characterised using a thermo-mechanical analyser (TMA), a differential scanning calorimeter (DSC), and a dynamic mechanical analyser (DMA). It was found that the storage modulus of the epoxy was increased in the presence of SS diffusants. However, the coefficient of thermal expansion (CTE) and the glass transition temperature ( $T_g$ ) of the epoxy ternary blends was reduced by the addition of SS diffusants, which was because the expansion of the epoxy matrix was constrained in the presence of silica fillers. The UV/Vis spectroscopy results demonstrated that the percentage of transmittance of epoxy was decreased by the incorporation of the silica diffusant.

**Keywords:** polymer composites, thermal properties, light-emitting diodes (LED), epoxy blends, silica

**Abstrak:** Adunan ternari epoksi (DCN) disediakan dengan pencampuran diglicidil eter bisfenol A (DGEBA), epoksi silkoalifatik, dan epoksi novolak. Difusan silika disediakan dengan penambahan silika sfera (SS) ke dalam adunan epoksi. Sifat-sifat terma bagi komposit epoksi dikaji dengan menggunakan penganalisis mekanik haba (TMA), kalorimetri pengimbasan pembezaan (DSC), dan penganalisis mekanik dinamik (DMA). Modulus simpanan bagi epoksi telah ditingkatkan dengan kehadiran difusan SS. Walau bagaimanapun, pekali pengembangan haba (CTE) dan suhu peralihan kaca ( $T_g$ ) bagi adunan ternari epoksi telah diturunkan dengan penambahan difusan SS disebabkan pengembangan matriks epoksi telah dihalang dengan kehadiran pengisi silika. Keputusan spektroskopi UV/Vis menunjukkan bahawa peratusan transmisi bagi epoksi dikurangkan dengan penambahan difusan silika.

**Kata kunci:** komposit polimer, sifat-sifat terma, diod pemancar cahaya (LED), adunan epoksi, silika

## 1. INTRODUCTION

Epoxy resins have become increasingly important because of the wide variety of their applications in the automotive, aerospace, electronics, and plastics industries, and because of their structural applications.<sup>1</sup> The typical characteristics of epoxy are good chemical and corrosion resistance, good mechanical and thermal properties, outstanding adhesion to various substrates, low shrinkage upon curing, high flexibility, good electrical properties, and the ability to be processed under a variety of conditions.<sup>2</sup> Epoxy is an important resin in the light emitting diode (LED) industry because it has good thermal stability and mechanical properties, and it is suitable for the encapsulation of silicon chips and lead frames.<sup>3</sup> Light emitting diodes have replaced incandescent, fluorescent, and neon lamps. The factors that make LEDs so common are due to their ability to produce high luminosity at low currents and voltages and match with silicon-integrated circuits. Besides, LEDs have low-power consumption, longer service lives, and are able to be transformed into different shapes.<sup>4</sup> LED lamps are encapsulated by transparent polymers such as epoxy resin with refractive indices in the range of 1.5–1.6.

In the LED industry, the most common epoxy resins are diglycidyl ether of bisphenol A (DGEBA) and cycloaliphatic epoxy resin (CAE). However, DGEBA epoxy resin tends to undergo discoloration; on the other hand, CAE is a brittle material.<sup>5,6</sup> Improvements have been done by many researchers to overcome the weakness of DGEBA epoxy resins and CAE. To solve this problem, DGEBA was blended with CAE to reduce the thermal discoloration. The blending of DGEBA with CAE could enhance the polymerisation rate even at low catalyst concentrations and, thus, subsequently reduce thermal discoloration.<sup>6</sup> According to Park *et al.*,<sup>7</sup> the blending of epoxy with other resins can be done so as to obtain better overall performance, such as ease of processing, good curing ability, high thermal stability, high chemical resistance, good mechanical strength, and good weathering. Kumar *et al.*<sup>8</sup> used different ratios of DGEBA/novolac/CAE in their study of ultraviolet radiation, curable epoxy resins encapsulants for LEDs. A blend of DGEBA with 10–50 wt% of epoxy novolac, derived from *p*-cresol, shows substantial improvement in elongation, the energy absorbed in order to break, and thermal stability.<sup>9</sup>

Generally, the addition of fillers could increase the thermal stability and thermal conductivity of epoxy. Besides, the incorporation of fillers could reduce the shrinkage, cost, and coefficient of thermal expansion.<sup>1</sup> Xu *et al.*<sup>10</sup> found that the addition of nano-silica could improve the toughness properties and thermal stability of CAE. Wazzan *et al.*<sup>11</sup> reported that the toughness and impact resistance of DGEBA epoxy increased by 60%–65% in the presence of 4 wt% of titanium dioxide. According to Haque *et al.*,<sup>12</sup> by dispersing 1 wt% of nanoclay, the shear strength, flexural strength, and fracture toughness of epoxy was

improved by 44%, 24%, and 23%, respectively. The use of silica in the world today is increasingly important. Silica is used in glass, coating, ceramics, paints, plastics, rubber, oil, electronics, and in the optical and construction industry.<sup>13</sup>

Light emitted from clear epoxy encapsulation is straight and focused. This light can only act like spotlight, and it is not suitable for daily use. Furthermore, the coefficient of the thermal expansion (CTE) for pure epoxy resin is very high. It is about 10 times higher than the CTE of a silicon chip and a lead frame. The CTE mismatch between the epoxy resin and the component in LEDs will induce internal thermal stress which is the main cause of epoxy encapsulation delamination. However, when a silica filler was added to the epoxy resin, the emitted light spread. This LED can act like an indicator for electronic equipment.<sup>14</sup> Therefore, a silica filler can be used to spread the emitted light and depress the CTE, and it has the lowest influence on emitted light transmittance. In the LED industry, silica particles were incorporated in epoxy resins to form a mixture called a diffusant. The diffusant was then mixed into the epoxy system for the LED encapsulation. The addition of the silica could reduce the CTE. Silica can be used to increase the dimensional stability, thermal conductivity, moisture content, and the electric and abrasion resistance of the material. In addition, the silica particle is relatively inexpensive.<sup>15</sup>

In this study, epoxy ternary blends were prepared by mixing DGEBA, cycloaliphatic epoxy and novolac epoxy. Attempts are made to investigate the effects of spherical silica (SS) on light transmittance, and the thermal and dynamic mechanical properties of the epoxy blends.

## **2. EXPERIMENTAL**

### **2.1 Materials**

The epoxy blends (DCN) were prepared by mixing DGEBA, cycloaliphatic epoxy and novolac epoxy at a predetermined ratio. The filler used in this study is SS. The specific surface area and average particle size of the SS is approximately 13 m<sup>2</sup>/g and 4 μm, respectively. Table 1 shows the epoxy equivalent weight (EEW) and the viscosity of liquid epoxy, DGEBA, cycloaliphatic epoxy, and novolac epoxy. Methylhexahydrophthalic anhydride (MHHPA) was used as a curing agent. The anhydride equivalent weight (AEW) of MHHPA is 168.2.

Table 1: EEW and viscosity of liquid epoxy, DGEBA, cycloaliphatic epoxy and novolac epoxy.

Epoxy resin	EEW	Viscosity at 25°C (cP)
Liquid epoxy	185–196	700–1100
DGEBA	185–192	1100–1500
CAE	131–143	350–450
Novolac epoxy	172–180	1100–1700

## 2.2 Preparation of Silica Diffusant

The silica diffusants were prepared by the addition of SS into liquid epoxy. The loadings of the SS into the liquid epoxy were 20%, 30%, 40%, and 50%. Thereafter, the diffusants were designated as SS20, SS30, SS40, and SS50, respectively. Firstly, the SS particles were dispersed into the liquid epoxy by using a mechanical stirrer at a speed of 1200 rpm for 1 hour. The mixture was then bottled and stirred by using an ultrasonic vibrator (Ultrasonik Ney 208H, USA) to reduce the size of the silica particle agglomerates.

## 2.3 Preparation of Epoxy/Silica Composites

For the preparation of unfilled epoxy, the epoxy ternary blends and the MHHPA (curing agent) were mixed in a ratio of 1:1. The mixture was cured at 110°C for 1 hour. The post-curing process was then carried out at 135°C for 2 hours in an oven. For the preparation of the epoxy/silica composites, the ratio between the epoxy resin and the MHHPA curing agent was set at 1:1. The epoxy ternary blends/silica diffusant mixtures were stirred by using a mechanical stirrer. The mixture was then poured into a silicon rubber mould. After that, the epoxy/silica composite was cured at 110°C for 1 hour followed by post-curing at 135°C for 2 hours in an oven. The percentage of diffusant in the epoxy blends was fixed at 4%, 8%, and 12%. The materials' compositions and designations are shown in Table 2.

Table 2: Materials' designations and compositions for the epoxy/silica composites.

Diffusant	Percentage of diffusant in epoxy blends (%)		
	4	8	12
SS20	DCN/SS20-4	DCN/SS20-8	DCN/SS20-12
SS30	DCN/SS30-4	DCN/SS30-8	DCN/SS30-12
SS40	DCN/SS40-4	DCN/SS40-8	DCN/SS40-12
SS50	DCN/SS50-4	DCN/SS50-8	DCN/SS50-12

## 2.4 Materials' Characterisations

### 2.4.1 Thermo-mechanical analysis

Thermo-mechanical analysis of the epoxy/silica composites was carried out by using a thermo-mechanical analyser TMA (TMA Diamond, Perkin Elmer, USA). The epoxy sample was heated from 30°C to 300°C at a heating rate of 5°C/min, in a nitrogen gas atmosphere. The height of the epoxy sample is in the range of 8–10 mm. The CTE and the glass transition temperature ( $T_g$ ) of the epoxy samples were calculated by using Pyris<sup>TM</sup> software (Perkin Elmer, USA).

### 2.4.2 Differential scanning calorimetry

Differential scanning calorimetry analysis was performed using a differential scanning calorimeter, DSC (Diamond analyser, Perkin Elmer, USA). The tests were carried out in a nitrogen gas atmosphere. For the uncured sample, a double scanning method was used. First, the sample was heated from 30°C to 250°C at a heating rate of 10°C/min in order to cure the sample. The sample was held at 250°C for 1 min. After that, the sample was cooled from 250°C to 30°C at a cooling rate of 20°C/min. The sample was then held at 30°C for 1 min. Second, a heating process that was similar to the first heating process was performed. The weight of the sample was in the range of 10–15 mg. The temperature at which the curing reaction began ( $T_{onset}$ ), the temperature at which the maximum curing reaction occurred ( $T_{peak}$ ),  $T_g$  and the enthalpy ( $\Delta H$ ) were all determined by using Pyris<sup>TM</sup> software. For the oven-cured sample, a single scanning method was used. The sample was heated from 30°C to 250°C at a heating rate of 10°C/min.

### 2.4.3 Dynamic mechanical analysis

Dynamic mechanical analysis was carried out by using a dynamic mechanical analyser (DMA 8000, Perkin Elmer, USA). The sample was heated from 30°C to 250°C at a heating rate of 2°C/min under a normal air environment. A single cantilever bending mode was performed on the epoxy samples. The vibration frequency was set at 1 Hz. The storage modulus ( $E'$ ), the loss modulus ( $E''$ ) and the  $T_g$  and were determined by using Pyris<sup>TM</sup> software.

### 2.4.4 Light transmittance tests

A UV/Vis spectrometer (Lamda 25, Perkin Elmer, USA) was used to measure the percentage of light transmittance for the epoxy/silica composite samples. Prior to the light transmittance test, a thin layer of the sample was cured on top of a glass plate. The emitted light's wavelength was set at the range of 300–1100 nm.

### 3. RESULTS AND DISCUSSION

#### 3.1 Thermo-mechanical Analysis

The CTE of the DCN/SS composites is shown in Figure 1. Note that the CTE of all of the DCN/SS composites is lower than that of the DCNs. It can be observed that the CTE of the DCN/SS composites was reduced as the content of the silica diffusant increased because silica exhibits a low CTE, at approximately 0.5 ppm/°C. As a result, this depresses the CTE of the silica-filled composites. A high amount of silica will then block the expansion of the DCN. It is believed that the expansion of the epoxy matrix will be constrained in the presence of silica fillers.<sup>16</sup> Similar observations were also reported by Wong *et al.*<sup>17</sup> and Chaturvedi and Shen.<sup>18</sup> Chaturvedi and Shen<sup>18</sup> studied the thermal expansion response of particle-filled polymer matrix composites using micro-mechanical modelling. It was found that the spatial distribution of filler particles plays a relatively small role in affecting the average composite CTE. However, the local stress field depends strongly on the particle arrangement.

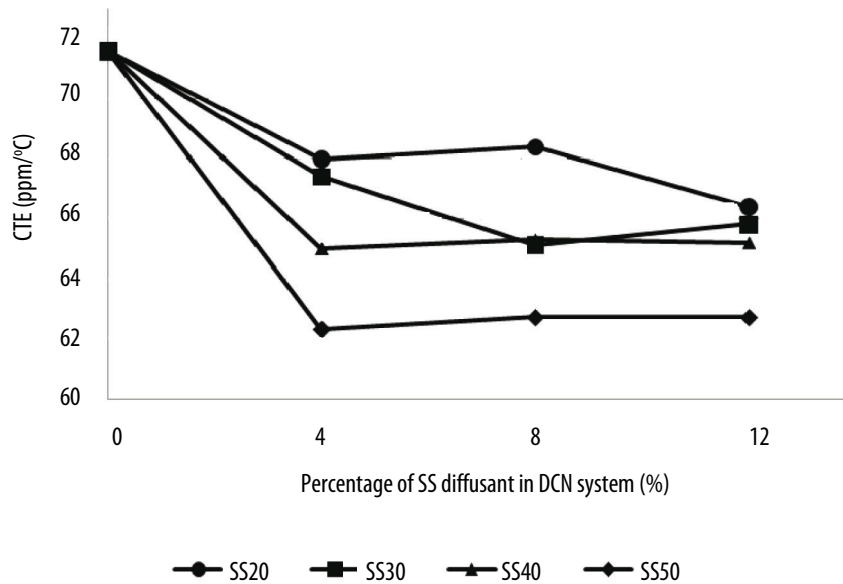
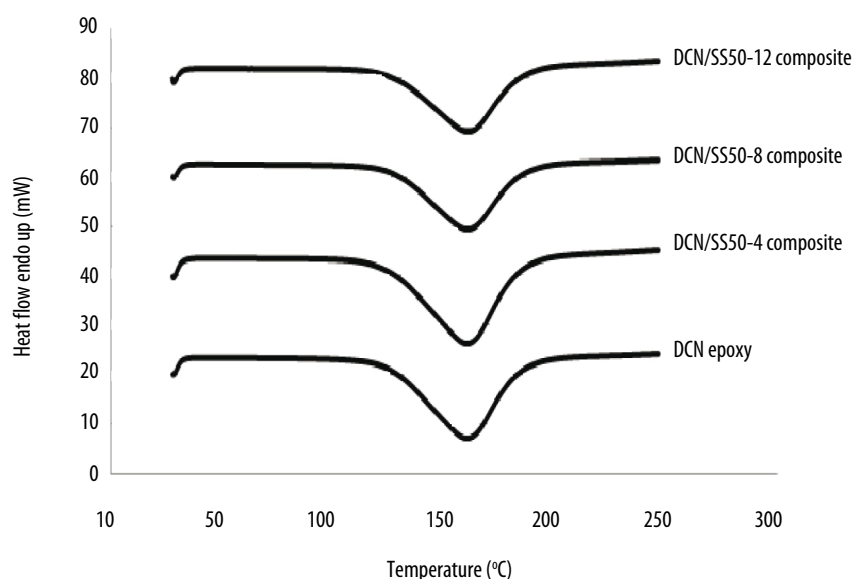


Figure 1: CTEs of various SS diffusant-filled DCN composites.

## 3.2 Differential Scanning Calorimetry

### 3.2.1 Curing reaction of uncured samples

The first and second DSC heating thermograms of the DCN/SS50 composites are shown in Figures 2(a) and 2(b), respectively. Table 3 shows the thermal characteristics, for example,  $T_{onset}$ ,  $T_{peak}$ ,  $T_g$  and  $\Delta H$ , of the DCN and the various DCN/SS composites. It can be seen that the  $T_{onset}$  of the epoxy blends increased with the addition of the silica diffusant, albeit the increment is not very significant. The  $T_{onset}$  of the DCN is 124.6°C. The highest  $T_{onset}$  was achieved by the DCN/SS50-8 composite, where the  $T_{onset}$  value was recorded to be 127.1°C. The variation between these 2 temperatures is only 2.5°C. Therefore, it is believed that the starting temperature of the cross-linking reaction remains unchanged by the addition of the silica diffusant. According to Liu *et al.*,<sup>19</sup> the onset point of the exothermic curve did not show an obvious shift when the colloidal silica was added to the epoxy system. The  $T_{peak}$  of the DCN was slightly increased by the incorporation of the silica diffusants. The  $T_{peak}$  of the DCN is approximately 160.6°C. The highest  $T_{peak}$  was achieved by the DCN/SS50-12 composite, where the  $T_{peak}$  value was recorded to be 161.7°C. This indicates that the SS diffusant-filled epoxy system still can be cured rapidly near a temperature of 160°C.



(a)

Figure 2: First (a) and second (b) DSC heating thermograms of the DCN and the DCN/SS50 composites (*continued on next page*).

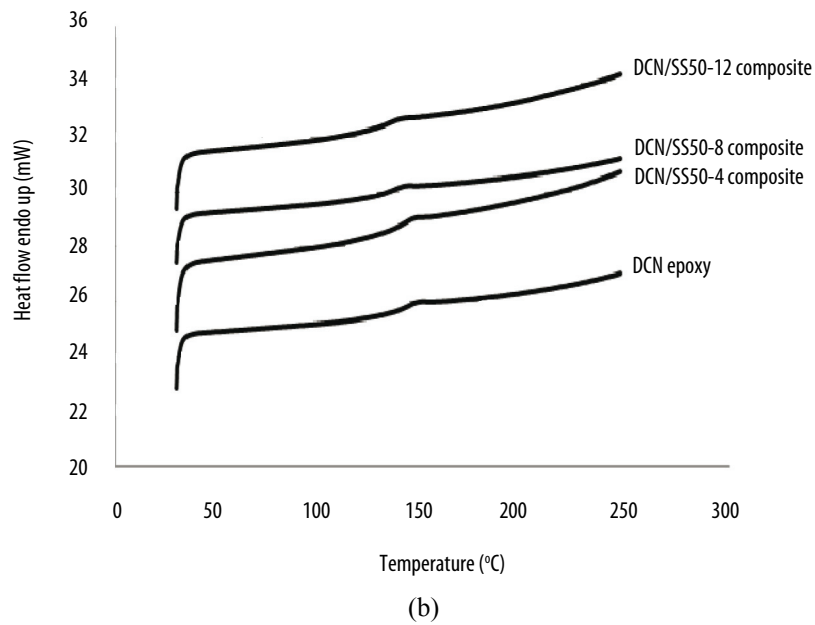


Figure 2: (continued).

Table 3: Thermal properties of the DCN and DCN/SS50 composites.

	<sup>a</sup> T <sub>onset</sub> (°C)	<sup>a</sup> T <sub>peak</sub> (°C)	<sup>a</sup> ΔH (J/g)	<sup>a</sup> T <sub>g</sub> (°C)	<sup>b</sup> E' at T <sub>g</sub> (GPa)	<sup>b</sup> E' at 30°C (GPa)	<sup>b</sup> T <sub>g</sub> (°C)
DCN	124.6	160.6	313.7	138.3	0.97	1.24	150.0
DCN/SS50-4	124.4	160.6	288.3	137.8	1.01	1.26	150.2
DCN/SS50-8	127.1	160.7	284.4	134.3	1.03	1.35	146.8
DCN/SS50-12	126.2	161.0	277.6	132.0	1.06	1.39	146.4

Notes: <sup>a</sup> – data recorded from DSC  
<sup>b</sup> – data recorded from DMA

The  $\Delta H$  of the DCN decreased with the addition of SS50 diffusants. The result suggests that the degree of cross-linking formation was depressed when the SS50 diffusant was added to the DCN. Note that the depression of cross-linking was higher with the increase in the loading of the diffusant. According to Macan *et al.*,<sup>20</sup> the incorporation of silica particles into the epoxy resin could decrease the formation of cross-linking. Table 3 shows that the  $T_g$  of the DCN/SS50 composite decreased as the percentage of the SS50 diffusant increased. This observation can be related to the degree of cross-linking in the epoxy system. The reduction of the degree of cross-linking could lead to the decrease of  $T_g$ . This again indicates that the SS particle could induce a reduction in the degree of cross-linking in the DCN. A similar observation was reported by Preghenella



*et al.*<sup>21</sup> They found that the  $T_g$  of the epoxy system decreased by the incorporation of fumed silica.

### 3.2.2 Curing reaction of oven-cured samples

The DSC heating thermograms of the DCN/SS50 composites are shown in Figure 2(b). The thermogram shows that an exothermic peak was revealed after  $T_g$ . This observation showed that the sample that was cured at 110°C for 1 hour followed by post-curing at 135°C for 2 hours by using an oven is a partially-cured epoxy system. This indicates that there are still a number of unreacted DCN and MHPA curing agents left in the sample after curing by the oven. Thus, cross-linking reactions are still able to occur during the DSC heat-scanning process. Figure 3 shows the changes in the  $T_g$  of the DCN when SS50 were added at different loading points. It can be seen that the  $T_g$  and the  $\Delta H$  of the DCN/SS50 composite decreased as the SS50 diffusant content increased.

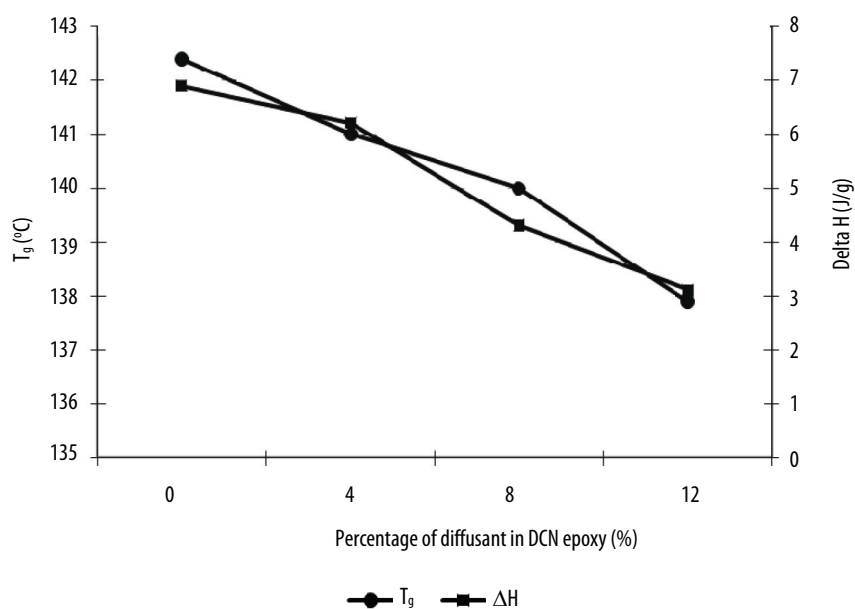
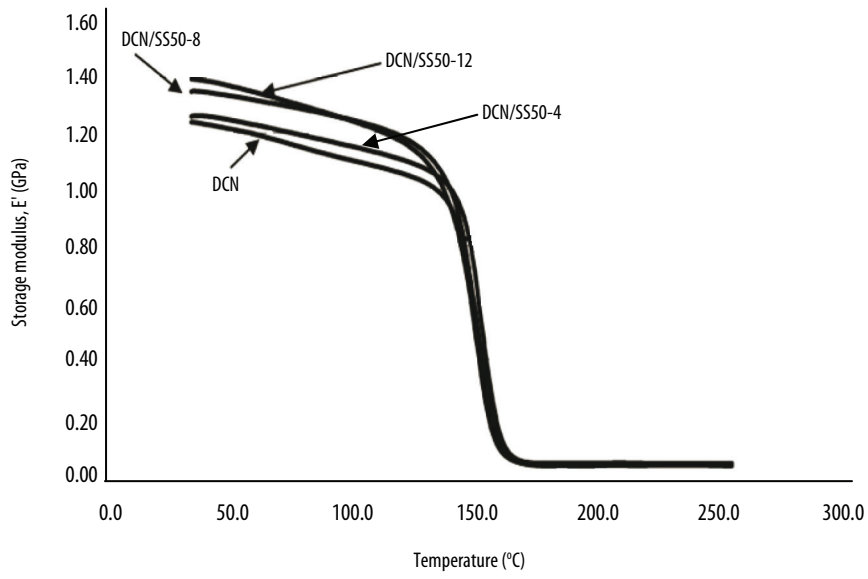


Figure 3: Effect of the percentage of diffusant in the DCN system on the  $T_g$  and  $\Delta H$  for the DCN/SS50 composites.

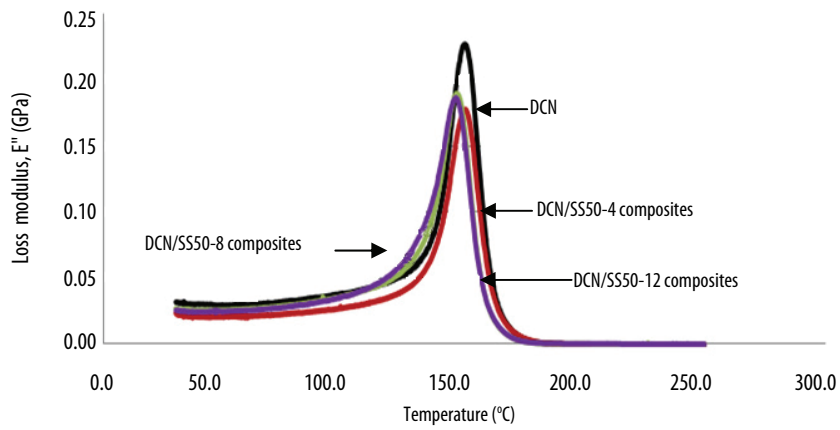
### 3.3 Dynamic Mechanical Analysis

According to Callister,<sup>22</sup> the magnitude of a thermal stress developed by a temperature change ( $\Delta T$ ) is dependent on the CTE and the  $E'$ . Thus, the thermal stresses will be reduced if the material contains low CTE and a low  $E'$ . The

dynamic  $E'$  for the DCN/SS50 composites as a function of the temperature is shown in Figure 4(a). Both the DCN and the DCN/SS50 composites showed typical visco-elastic behaviour: a glassy region, a glass transition region, and a rubbery region. As shown in Figure 4(a), the  $E'$  in the glassy region decreased as the temperature increased. This phenomenon was due to the CTE difference between the silica particle and the epoxy resin. The value of  $E'$  at 30°C is similar to the  $E'$  at room temperature. In the LED industry,  $T_g$  is the upper limit of service temperature. After  $T_g$ , the LED lens will become rubbery, and will malfunction. Therefore,  $E'$  at 30°C and a  $T_g$  were chosen for further comparison. Table 3 shows the  $E'$  of the DCN and the DCN/SS50 composites recorded at a temperature of  $T_g$  and at 30°C. The  $E'$  for the DCN/SS50 composites was higher than that of the DCN epoxy. This is attributed to reinforcement by silica particles. Adachi *et al.*<sup>23</sup> reported that the silica particles increase the  $E'$  of the epoxy/silica composites. The  $E'$  of the DCN/SS50 composites increased gradually as the percentage of SS50 diffusants increased. This might be associated with the strong interaction between the SS particles and the DCN. Wong *et al.*<sup>17</sup> reported that the strong interaction between the silica particle and the epoxy matrix could lead to an increment of the  $E'$  for the composite. The  $E''$  for the DCN/SS50 composites as a function of temperature is shown in Figure 4(b). Figure 4(b) shows that the height of  $E''$  decreased as the percentage of the SS50 diffusant increased. Ragosta *et al.*<sup>24</sup> observed that the peak of  $E''$  of the epoxy/silica composite decreased with the addition of the silica fillers. They suggested that this may be due to the non-dissipative nature of the filler which reduces the visco-elastic response of the composites. Note that the  $E''$  peak shifted to a lower temperature as the SS50 diffusant contents increased. The shifting is due to the depression of  $T_g$  as the content of the SS50 diffusant increases. Table 3 shows the  $T_g$  of the DCN and of the various DCN/SS50 composites. The  $T_g$  for the DCN/SS50 composites slightly decreased as the percentage of SS50 diffusant increased. The difference in the  $T_g$  value between the DCN and the DCN/SS50 composites was less than 5%. These results are in line with the data obtained from the DSC, as discussed in the earlier section. The degree of cross-linking decreased as the filler content increased. Thus, the reduction of  $T_g$  might be due to the depression of the cross-linking density.



(a)

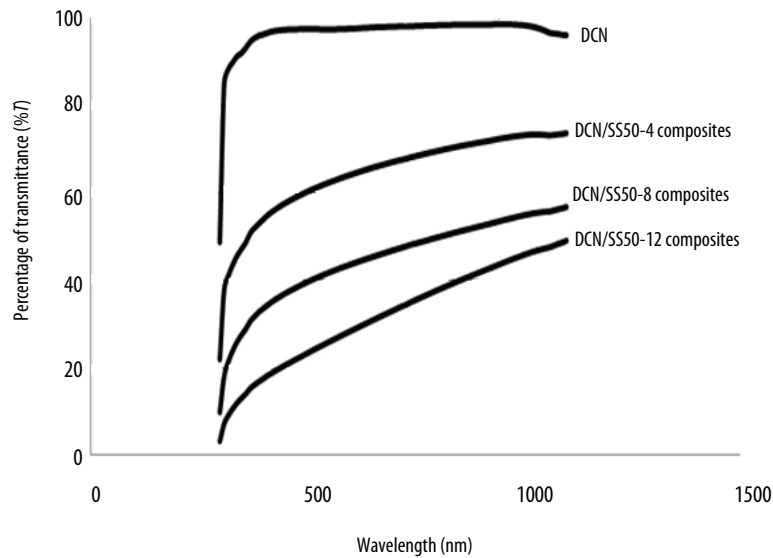


(b)

Figure 4: Dynamic storage modulus ( $E'$ ) (a) and loss modulus ( $E''$ ) versus temperature (b) for DCN and DCN/SS50 composites.

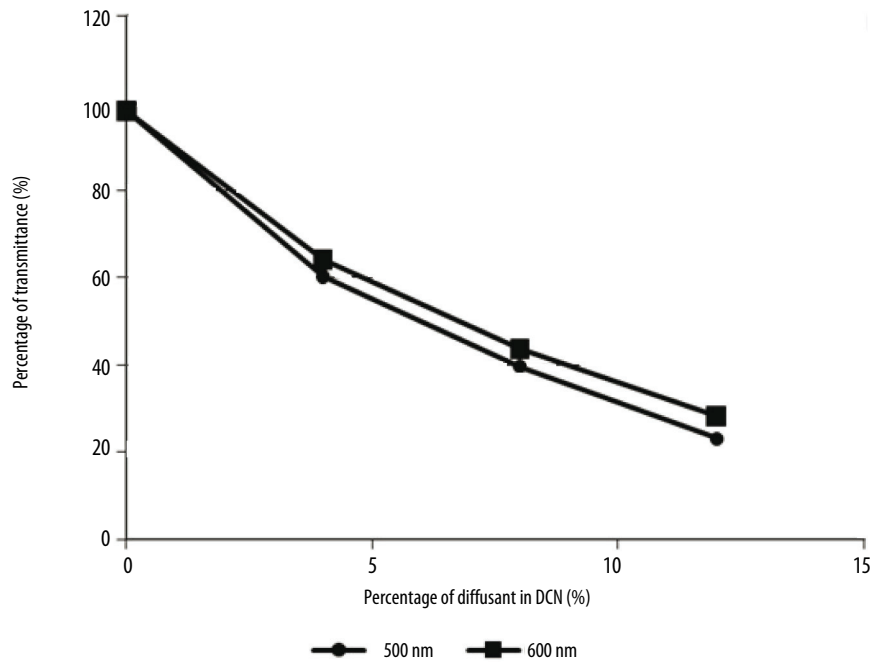
### 3.5 Light Transmittance Tests

The light transmittance of the DCN/SS50 composites as a function of wavelength is shown in Figure 5(a). The percentage of light transmittance (% $T$ ) for the DCN and DCN/SS50 composites appears to be above the wavelength 300 nm. After 300 nm, the % $T$  increased as the wavelength increased. This is a natural phenomenon because the molecular level of light absorption of an epoxy and glass appears at a wavelength of shorter than 300 nm. Similar observations were also reported by Naganuma and Kagawa.<sup>25</sup> Figure 5(b) shows the % $T$  at 500 nm and 600 nm for the DCN and the DCN/SS50 composites. It can be seen that the % $T$  of the DCN/SS50 composites decreased as the percentage of diffusants for SS50 increased. According to Kagawa *et al.*,<sup>26</sup> the % $T$  for the epoxy decreased as the glass fibre content increased. From Figure 5(b), one may observe that the trend for the % $T$  taken from 600 nm for the SS50-filled DCN is similar to the % $T$  data that were recorded at 500 nm. Sato *et al.*<sup>27</sup> studied the light transmittance of glass particle-dispersed epoxy matrix optical-composites, and it was found that the light transmission spectrum of the epoxy composites could be correlated with the wavelength dispersion curves of the refractive indices of the glass particles and the matrix.



(a)

Figure 5: Percentage of transmittance (% $T$ ) as a function of wavelength (a) and at 500 nm and 600 nm (b) for DCN epoxy and DCN/SS50 composites (*continued on next page*).



(b)

Figure 5: (continued).

#### 4. CONCLUSION

Based on this work and studying the effect of SS diffusants on the dynamic mechanical thermal properties and light transmittance of epoxy, the following conclusions can be drawn:

The CTE of the DCN/SS composites are lower than the CTE of unfilled DCN. Among all of the DCN/SS composites, it was found that the DCN/SS50 composite possesses the lowest CTE value. The  $T_{onset}$  and  $T_{peak}$  of the DCN were not influenced by the addition of diffusants. On the other hand, the  $\Delta H$  and  $T_g$  of the DCN decreased with the addition of SS50 diffusants. The DCN/SS50 composite exhibits improved the storage modulus by increasing the percentage of the SS50 diffusants. However, the % $T$  of the DCN/SS50 composites decreased as the percentage of SS50 diffusants increased. The light transmittance of the DCN/SS50-4 composites could achieve approximately 60% in the wavelength of 500 and 600 nm. Hence, a balance of the CTE,  $T_g$ , and the % $T$  of the epoxy blends/SS could be selected based on their required performance for the LED application.

## 5. ACKNOWLEDGEMENT

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