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# Enhanced Thermal Conductivity and Mechanical Toughness of the Epoxy Resin by Incorporation of Mesogens Without Nanofillers

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**ABSTRACT** Epoxy resin is widely used due to its electrical insulation performances, but low elongation defects at the break, low thermal conductivity, and high brittleness limit its application scenarios. In this paper, the mesogen is utilized to improve the thermal conductivity and mechanical toughness of the traditional epoxy resin. Both the mechanical performances, including the impact strength, tensile strength, the bending strength, and the thermal behaviors, including the thermal conductivity, are investigated. Results show that with the biphenyl liquid crystal epoxy resin (BLCER) content of 10%, the impacting strength, tensile strength, and bending strength are increased by 71%, 21%, and 11%, respectively. The thermal conductivity of the composites increases to 2.26 times that of pure epoxy resin. Both the enhanced mechanical and thermal performance of the epoxy resin by the mesogen incorporation are further investigated. It is indicated that the mesogen in the Biphenyl Liquid Crystal Epoxy Resin significantly improve both the mechanical toughness and thermal conductivity of the epoxy resin by the formation of the micro-crack behavior and the thermal conductive networks, respectively. With the aids of the mesogen, the improved variety of properties in epoxy resin without reducing its original performance is attractive in the industry application with great demand in the balance of the comprehensive performances.

**INDEX TERMS** Mesogen, epoxy resin, toughness, thermal conductivity.

## I. INTRODUCTION

Epoxy resin-based composites widely utilized due to their excellent electrical insulation and chemical resistance are able to protect the electrical equipment from insulation breakdown, short-circuit, electrothermal chemical aging, and electric field distortion [1]–[5]. Epoxy resin exhibits strong performance versatility, thermochemical stability under severe conditions, and convenient processing. Thus, the high electrical insulation strength of the epoxy resin has significantly improved the reliability of the electrical equipment [5], [6]. To further improve the thermal conductivity, mechanical toughness performances of epoxy resin-based composites, and make them suitable for more cutting-edge fields, epoxy resin blends with inorganic fillers have been widely studied [7], [8]. However, epoxy resin with a high degree of cross-linking will normally enhance its thermal

conductivity at the cost of reduced mechanical properties [9]. In both performances, contradictions cannot meet the requirements in the heat dissipation and structured stability of large power equipment in the power system.

Rigid nanoparticles can absorb the crack energy during the deformation process. Nanofillers such as the Al<sub>2</sub>O<sub>3</sub>, TiO<sub>2</sub>, and SiO<sub>2</sub> can be used as fillers to toughen the epoxy resin [10]–[13]. The reduced crack propagation energy avoids the generation of destructive cracks to achieve the purpose of toughening epoxy resin. Still, the toughened hybrids make the rheological performance poor and affect its processing and final shaping [14]. Inorganic fillers sometimes disperse unevenly, causing agglomeration to aggravate rigidity and brittleness of the composites [15], [16].

The combination of organic resin and epoxy resin not only enhance the performance of the resin itself but also avoid some negative effects brought by inorganic fillers. For this reason, scholars have carried out studies on the fracture toughness of epoxy resins and tried to use various

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toughening or reinforcing agents in order to improve the physical properties. The most traditional method is to modify the epoxy resin with rubber elastomers [17], [18]. Elastomers such as nitrile rubber and polyurethane can greatly improve the toughness of epoxy resins, but it sometimes reduces its heat resistance and affects the thermal conductivity. The toughening agent IPNs and epoxy resin form an interpenetrating network IPN polymer system that can greatly improve the toughness of epoxy resin without reducing its thermal stability and tensile strength [19], [20]. Still, the thermal stability and other physical properties will be damaged by factors such as compatibility, dispersion rate of active groups, etc. Hyperbranched polymer (HBP) is featured with good compatibility with epoxy resin and high curing efficiency [21], [22]. It can be toughened without destroying the performance of epoxy resin itself, but hyperbranched fillers will be twisted around, resulting in that it can't be applied to the scene. Zhang *et al.* reported the effect of liquid crystal epoxy resin content on the curing speed and mechanics of epoxy resin [23]. However, the improvement in both the thermal conductivity and the mechanical performances of the epoxy resin without inorganic nanofillers is rarely reported. The fundamental issue of the modified epoxy are cooling problems and mechanical toughness problems. The cooling problem is that the miniaturization and high power of electronic devices will generate a lot of heat and the voltage level and capacity of large power equipment are increasing. And the mechanical toughness problem is about the cured epoxy resin has a highly cross-linked network structure, which will weaken the ability of brittleness and impact strength and the ability of stress crack.

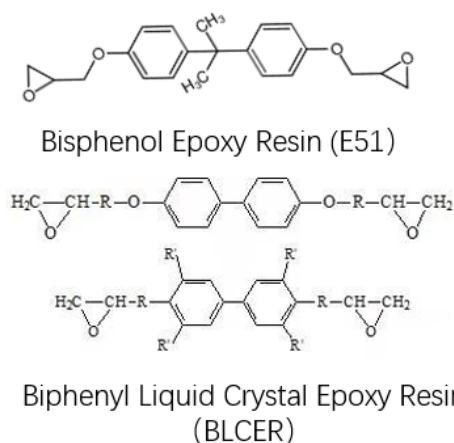
In this paper, the mesogen is blended with epoxy resin by introducing liquid crystal resin into the epoxy resin matrix. The influence of the liquid crystal epoxy resin contents on the mechanical properties, thermal stability, and thermal conductivity of the mesogens/epoxy resin composites is studied. Furthermore, the enhancement in the toughening and thermal conductivity performances of epoxy resin composites is analyzed.

## II. MATERIALS AND METHODS

### A. MATERIALS

The epoxy equivalent of the biphenyl liquid crystal epoxy resin (BLCER) is 265-285 g/mol. The light yellow and spherical solid-state BLCER is got from the Gansu Institute of Chemical Industry as received. Both of bisphenol epoxy resin(E-51) with an epoxy equivalent 182-192 g/mol, and the curing agent of 4,4'-diamino diphenyl sulfone (DDS) with an epoxy equivalent of 248.3 g/mol and purity of more than 97%, were obtained from Aladdin Reagent Co. All the reagents are analytically pure. Figure 1 shows the molecular structures of BLCER and bisphenol epoxy resin(E-51).

The addition of liquid crystal epoxy resin improves the thermal stability of the cured product. After adding liquid crystal epoxy resin, the initial decomposition temperature (weight loss 5%) of the cured product increased by 21.8 °C



**FIGURE 1.** Molecular structure of bisphenol epoxy resin(E51)and molecular structure of biphenyl liquid crystal epoxy resin(BLCER).

to 394.5 °C. This is due to the longer rigid segments in the mesogenic unit in the liquid crystal epoxy, and the tight arrangement of the molecules in the liquid crystal region, which reduces the free volume in the cured epoxy resin network and increases the crosslink density of the cured product, so the thermal stability becomes better. Similarly, after adding liquid crystal epoxy resin, the weight loss temperature of the curing system is also increased. When adding liquid crystal epoxy resin, the temperature of weight loss increased by 12.2 °C.

### B. SAMPLE PREPARATION

The weight ratios between the liquid crystal epoxy resin and conventional epoxy resin were shown in Table 1. The solid BLCER was heated to a liquid state with a temperature of 180 °C. 100 g of epoxy resin was added to a 500 ml beaker at the same time. To reduce its viscosity, the epoxy resin was heated to 70 °C. An appropriate amount of liquid crystal epoxy resin was poured in a molten state, with further heating up to 100 °C and mechanical stirring. The hardening agent 4,4'-diamino diphenyl sulfone was gently stirred until it was completely melted. The mixture was moved to a vacuum-drying oven for 20 minutes to remove air bubbles, followed by a solidifying process for 2 hours under the temperature of 180 °C in the preheated mold. After curing, it was slowly cooled down to room temperature and de-molded to obtain BLCER/E-51/DDS composites with different BLCER contents.

**TABLE 1.** The weight ratios between the liquid crystal epoxy resin and pure epoxy resin.

Identification	E-51/wt%	BLCER/wt%
0#	100	0
1#	98	2
2#	95	5
3#	90	10
4#	80	20

### C. TESTING AND CHARACTERIZATION

#### 1) DETERMINATION OF IMPACT STRENGTH

The impact tester(ZBC2302-4) was used to determine the impact resistance of the resin composites. The sample was prepared according to GB/T2567-2008, whose size was 80 mm × 10 mm × 4 mm, and there was no initial notch impact. The calculation formula is:

$$ak = \frac{A}{b \cdot h} \quad (1)$$

where  $a_k$  represents the impact strength in  $\text{kJ/m}^2$ ,  $A$  is the energy consumed by the impact sample in MJ whereas,  $b$  indicates the width of the sample in mm, and  $h$  is the thickness of the sample in mm.

The test is performed at room temperature, and each group of samples is tested five times. The average value is taken as the impact strength of the samples.

#### 2) DETERMINATION OF TENSILE STRENGTH

The universal electronic tester(AG-X10KN) was used to determine the tensile strength of the composites, and samples with a thickness of 1 mm were prepared following GB/T2567-2008. The stretching rate is 25 mm/min. The calculation formula is:

$$T = \frac{P}{b \cdot h} \quad (2)$$

where  $T$ ,  $P$ ,  $b$ , and  $h$  represent the tensile strength in MPa, the breaking load in N and the width of the sample in mm, and the thickness of the sample in mm, respectively. The sample testing is performed at room temperature, and each group of samples is tested five times. The average value tested is considered as the tensile strength of the samples.

#### 3) DETERMINATION OF MATERIAL BENDING STRENGTH

The universal electronic tester(AG-X10KN) is used to measure the bending strength of the composites. The size of the sample strip is in 10 mm × 8 mm × 4 mm. The testing span and the moving speed is 20 mm and 2 mm/min, respectively. In this experiment, a three-point bending method was used to test the bending strength of the composite. The calculation formula for experimental bending strength ( $\sigma_f/\text{MPa}$ ) is as follows:

$$\sigma_f = \frac{3PL}{2bd^2} \quad (3)$$

where  $\sigma_f$  corresponds to the bending strength in MPa.  $P$  represents the specimen breaking in N.  $L$  is the distance between fulcrums (span) in cm. The  $b$  and  $d$  are the width and thickness of the specimen in cm, respectively. The average value is regarded as the impact strength of the group.

#### 4) SECTIONAL SCANNING ELECTRON MICROSCOPE (SEM) TEST

The cross-section of the mesogen modified epoxy resin after the impact test is cleaned with the acetone reagent. The micro-morphology of the cross-section of the sample was observed

with SEM(Hitachi, Japanese) with an accelerating voltage of 25 kV.

#### 5) THERMOGRAVIMETRIC ANALYSIS (TGA)

The thermogravimetric analysis curve of the BLCER/E-51/DDS blends with different BLCER contents was tested with the TGA-1 thermogravimetric analysis from Mettler Company. The BLCER/E-51/DDS blends with different BLCER contents were weighted about 5 mg, and the gas flow rate under a nitrogen atmosphere was kept at 50 mL/min. The sample heating temperature was ranged between 30 °C and 800 °C. And the heating rate was kept at 10 °C/min.

#### 6) THERMAL CONDUCTIVITY TEST

The thermal conductivity of BLCER/E-51/DDS blends was measured by the laser thermal conductivity meter(NETZSCH LFA457, German). The samples are in the form of small discs with a diameter of  $12.70 \pm 0.10$  mm and a thickness of  $1.00 \pm 0.10$  mm. The surface of the sample is kept flat and free of defects. The test is performed at room temperature with a laser voltage of 1.6340 kV.

The thermal conductivity at room temperature is determined by the coefficient of thermal expansion  $\alpha$ , the density  $\rho$ , and the total specific heat capacity  $C_p$ . The following formula calculates the thermal conductivity  $\gamma$ :

$$\gamma = \alpha \rho C_p \quad (4)$$

The coefficient of thermal expansion  $\alpha$  is based on the measurement from the laser meter of thermal conductivity. Before the measurement, it is necessary to spray a layer of graphite coating on the prepared sample. This is to increase the thermal contact and to prevent the laser from directly passing the sample itself. The density  $\rho$  is measured by the drainage method, whereas the specific heat capacity  $C_p$  is measured by a differential scanning calorimeter.

## III. RESULTS AND DISCUSSION

### A. THE PERFORMANCES OF THE MESOGEN BLENDED EPOXY RESIN

#### 1) MECHANICAL PROPERTIES

Figs. 1, 2, and 3 reflect the influence of different BLCER contents on the mechanical properties of the BLCER/E-51 blends. It is obvious from the testing that the tensile, flexural, and impact strength of the BLCER/E-51 blends increase to a certain extent, with the aids of the BLCER.

It can be seen from Fig. 1 that the increase of BLCER contents can enhance the impact strength of the BLCER/E-51 blends. When the BLCER content is less than 15%, the impact strength of the blend increases with the addition of BLCER. A maximum of 40.1  $\text{kJ/m}^2$  in the impact strength was reached with the aids of 20% of BLCER, which is 1.73 times that of the pure E-51. Fig. 2 shows the tensile strength of the BLCER/E-51/DDS blends. The tensile strength of the epoxy resin/mesogen blend is increased by 17 MPa with the BLCER content of 20%. The bending strength of the BLCER/E-51/DDS blend also shows

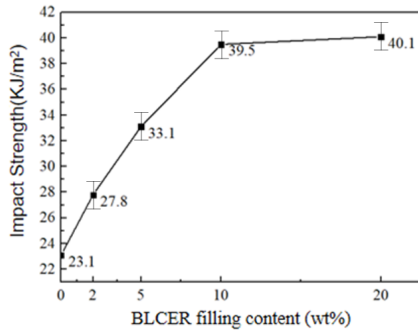


FIGURE 2. The relationship between the impact strength and the contents of BLCER of the blends.

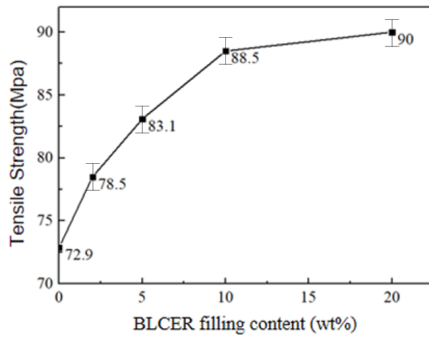


FIGURE 3. The influence of the contents of BLCER on the tensile strength of the blends.

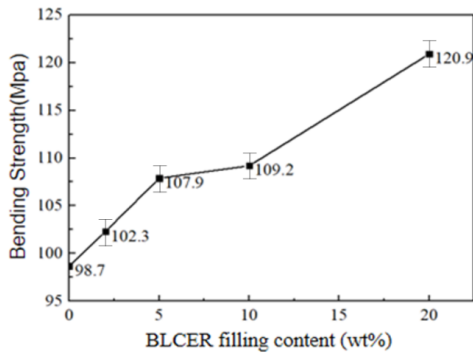


FIGURE 4. The impact of the contents of BLCER on the bending strength of the blends.

a significant increase by 22%, compared with that of the pure epoxy resin, as shown in Fig. 3.

2) THERMAL PROPERTIES

(1) Thermal stability

Under a nitrogen atmosphere, thermogravimetric analysis of BLCER/E-51/DDS blends with different BLCER contents was performed. Fig. 5 shows the thermogravimetric curve of the blends when the BLCER content is between 0% and 20%. The TGA testing results of different BLCER contents in the resin blends are listed in Table 2. The similar TGA curve of the BLCER/E-51/DDS blend with that of the pure E-51 indicates that all resin systems have similar thermal decomposition mechanisms. On the other hand, the addition of BLCER into the blends significantly increases the thermal

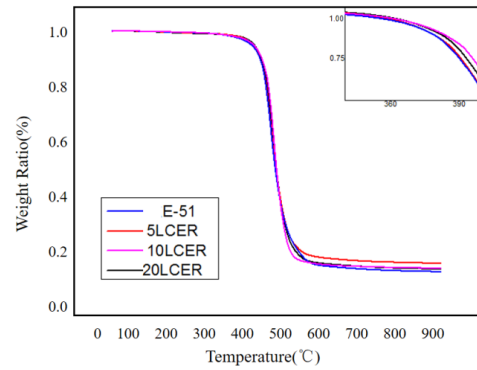


FIGURE 5. TGA curves of the resin blends with different BLCER contents.

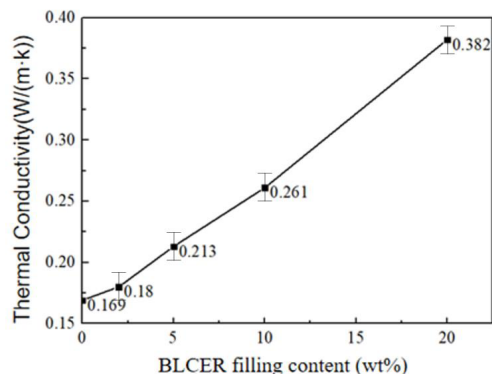
TABLE 2. TGA data of different BLCER contents in the resin blends.

Quality score (%)	Weightlessness temperature(°C)	
	5%	50%
0	349.33	405.30
5	354	406.77
10	355.33	408.67
20	357.31	409.67

decomposition temperature of the blends. When the BLCER content is increased to 20%, the initial decomposition temperature of the cured product (5% of the weight loss) increases by 7.92 °C. The formation of crystalline regions in the network produces a positive synergistic effect, which increases the cross-linking density of the blends, thereby improving the thermal stability of the blends. Besides, the addition of BLCER may increase the effect of hydrogen bonding between molecules during the curing process, thereby also improving the thermal stability of the blends.

3) THERMAL CONDUCTIVITY

Fig. 6 shows the effect of different BLCER contents on the thermal conductivity of the BLCER/E-51/DDS blends. As the BLCER contents are increased, the thermal conductivity of the blends is also increased. When the BLCER content is increased to 20%, the thermal conductivity of the blends is increased to 0.382 W/(m·K), which is 2.26 times greater than that of the pure E-51. The mesogenic units dispersed in the cross-linking matrix maintain the orientation structure of the molecules. When the BLCER content is high, the mesogenic units of BLCER dispersed in the E-51 matrix form a heat conduction path through a mutual contact. The heat passed through one mesogen unit is transferred to the next mesogen unit in contact with it, thereby effectively suppressing the phonon scattering and improving the heat transfer rate of the blends. On the other hand, the addition of BLCER in the blends also makes the structure of the blending system more compact in order, thereby reducing the phonon scattering caused by some voids in the cross-linked network of the



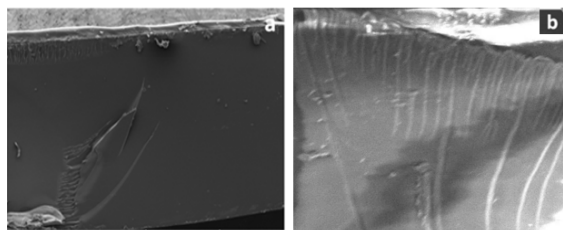
**FIGURE 6.** The effect of the BLCER content on the thermal conductivity of the blends.

amorphous region and also contributing to the improvement of the thermal conductivity of the blends.

**B. THE MECHANISM INVESTIGATION OF THE MESOGEN BLENDED EPOXY RESIN**

**1) SEM IMAGES OF THE FRACTURE SURFACES**

Fig. 7 shows the SEM images of the impact fracture of the blends with different contents of mesogen. The cross-section SEM image of the pure E-51 is featured with a typical brittle fracture with a smooth surface and almost no stripes. The fracture spreads out on the same plane without hierarchical features. Fig. 7(b) shows the fracture surface after mesogen modification. The fracture on the mesogen blended epoxy resin is uneven, with a large number of dimples and obvious micro-cracks on it. The scattered and staggered crack propagation pulls out many staggered fibrous structures, which are mesogenic domains with orderly orientation. The mesogen in-situ strengthens and toughens the epoxy resin blends. The mesogen blended epoxy resin presents a macroscopic two-phase structure, and there is a certain adhesion between the two phases, showing the characteristics of ductile fracture.



**FIGURE 7.** Scanning electron micrographs of the impact fracture of the blends with different mesogen contents: (a) pure E-51, (b) 20% of BLCER in the blends.

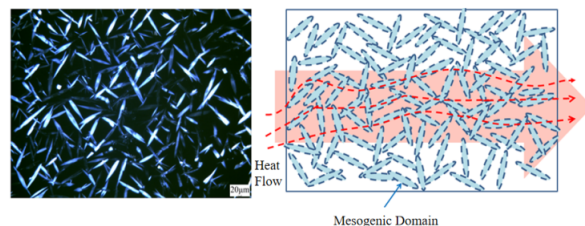
**2) TOUGHENING MECHANISM OF MESOGEN BLENDED EPOXY RESIN**

In the solidification of BLCER/E-51/DDS molten blend, the rigid rod-shaped liquid crystal molecules in BLCER are subjected to the shear force generated by the solidification contraction of the epoxy resin, and the orientation of mesogen molecules results in an ordered structure in the direction of the shear force. The cross-linking network formed by

the curing of epoxy resin fixed the microfibrillar domain. Simultaneously, the filamentous microfiber structure can be seen from the cross-section scanning electron microscope, which proves the existence of a rigid rod-like mesocrystalline domain in epoxy resin. The polyphase structure was thus formed in the BLCER/E-51/DDS blend. This polyphase structure can also be seen in polarized light microscope photographs. Observation under the polarized light shows that the mesocrystalline in the mesocrystalline domain are arranged in a directed and ordered manner. Due to the difference in mechanical strength and stiffness between the mesogen and epoxy resin, stress concentration will occur at the interface of the mixture after external impact. When cracks are developed in the composites, the mesocrystalline domains can avoid the further development of cracks and thus improve the toughness of epoxy resin. The more the microfibril domain in epoxy resin, the better the mechanical properties of epoxy resin blends.

**3) THERMAL CONDUCTIVITY MECHANISM OF LIQUID CRYSTAL EPOXY RESIN MODIFIED EPOXY RESIN**

The influence of the above different mesogen contents on the thermal conductivity of the BLCER/E-51/DDS blend indicates that the addition of liquid crystal epoxy resin can improve the thermal conductivity of ordinary epoxy resin. The introduction of a highly ordered mesogen can promote phonon transmission in the ordinary epoxy resin. Specifically, the highly ordered structure of the crystal phase can increase the mean free path of phonons, which is beneficial to the improvement of thermal conductivity. As shown in FIG. 8, compared with pure E-51, the mesogen blended epoxy resin demonstrates rigid rod-like liquid crystal microdomains, dispersed in the matrix with the oriented structure. With the increase of mesogen content, the mesocrystalline units of BLCER contact with each other again, forming a heat conduction path in the E-51 matrix.



**FIGURE 8.** Schematic diagram of heat transfer of mesogen blended epoxy resin composites.

**IV. CONCLUSION**

In this paper, by reacting different proportions of molten biphenyl liquid crystal epoxy resin (BLCER), ordinary epoxy resin(E-51), and curing agent, the BLCER/E-51/DDS blend with different BLCER content can be concluded as follows:

- (1) When the BLCER content is added to 20%, the thermal conductivity is 0.382 W/(m·K), which is 2.26 times higher than that of the pure E-51.

(2) Ordinary epoxy resin E-51 matrix shows a homogeneous disordered structure. Still, with the increase of BLCER content, the microstructure of the modified thermosetting material shows a transition from disordered structure to the ordered structure of rigid rod-like liquid crystal molecules. The thermal conductivity of the modified epoxy resin composite has also increased from 0.16 W/(m·K) to 0.38 W/(m·K).

(3) The BLCER/E-51/DDS blends is a partially compatible system, and rigid rod-shaped mesogenic units are formed in the cured product. The size and dispersion density of the mesogenic elements in the blends increases with the BLCER content. At the same time, the thermal conductivity of the composites also increases.

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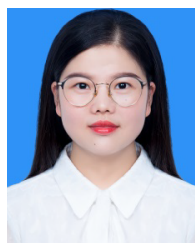
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