

## Hexagonal Boron Nitride (hBN) and Cordierite in Epoxy Underfill Materials: Single Filler System

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### ABSTRACT

As electronic devices become increasingly powerful and compact, the materials used for packaging must withstand high thermal and mechanical stress. Epoxy underfill materials are essential in providing thermal conductivity, mechanical reinforcement, and electrical insulation. However, differences in thermal expansion among silicon chips, solder joints, and substrates can cause failures over time. This research investigates the enhancement of epoxy composites through the addition of hexagonal boron nitride (hBN) and cordierite fillers to improve their mechanical, thermal, and dielectric properties. Composites were prepared with filler loadings ranging from 0 to 9 vol%, utilising mechanical stirring and ultrasonic dispersion, with diethyl toluene diamine (DETDA) serving as the hardener. The study evaluated properties such as viscosity, flexural strength, fracture toughness, thermal conductivity and coefficient of thermal expansion (CTE), thermal stability, and dielectric behavior. Testing was performed using a Universal Testing Machine (UTM), Scanning Electron Microscope (SEM), KD2 Pro thermal analyzer, Linseis L75 dilatometer, NDJ-9S viscometer, and an LCR meter. Results indicated that hBN fillers significantly increased thermal conductivity, reaching up to 0.340 W/m·K at 9 vol% loading, while cordierite fillers improved mechanical strength, achieving a maximum flexural strength of approximately 119.91 MPa. Higher filler contents led to increased viscosity, with hBN composites demonstrating notably higher viscosity than cordierite-based systems. These findings suggest that hBN is more suitable for thermal management, whereas cordierite offers better structural reinforcement. Overall, the study highlights the critical role of filler selection in customising epoxy composites for advanced electronic packaging applications.

**Keywords:** Cordierite, Dielectric Properties, Epoxy Composite, Hexagonal Boron Nitride, Thermal Conductivity

### 1. INTRODUCTION

In recent years, managing heat dissipation has emerged as a critical challenge in high-density and high-power electronic devices, driven by the rapid miniaturization of microelectronic components [1]. Epoxy is widely used as the base matrix in electronic packaging materials due to its excellent adhesion, mechanical strength, and thermal stability, which are essential for enhancing the reliability of electronic packaging [2]. To enhance their performance, these epoxy matrices are often reinforced with functional fillers that can improve thermal conductivity, mechanical robustness, and dielectric properties [3]. Among the promising filler options are hexagonal boron nitride (hBN) and cordierite, each offering unique advantages. hBN is notable for its high electrical resistivity [4], thermal conductivity [5], mechanical strength, and chemical and thermal stability, along with a lightweight nature. Conversely, cordierite provides high tensile strength, a low dielectric constant, superior thermal stability, and a very low coefficient of thermal expansion [6]. These characteristics make cordierite particularly suitable for

reducing thermally induced stresses and maintaining dimensional stability in packaging materials. This paper aims to examine the performance of two fillers, hexagonal boron nitride (hBN) and cordierite, when used alone as an epoxy underfill system. This paper will determine the best vol.% of hBN and cordierite to improve epoxy underfill by considering different vol.% of both substances.

### 2. THEORETICAL BACKGROUND

#### 2.1. Single Filler System in Epoxy-Based Underfill Materials

Epoxy resins are generally preferred as the material of the matrix in microelectronic packaging because of their high adhesion, chemical resistance, thermal stability, and dielectric insulating properties [7]. However, pure epoxy resins normally lack good thermal conductivity (~0.2 W/m·K) and have a large coefficient of thermal expansion (CTE). This can cause a thermal mismatch between the substrate and the chip. As a result, delamination or device failure will

occur during thermal cycling [8]. Thus, thermostable fillers are added to epoxy to improve the performance of the materials. There is one type of filler that is suitable to be used because of its impact on thermal, mechanical, and dielectric behaviours. Hexagonal boron nitride (hBN) is selected because it has higher thermal conductivity (up to 600 W/m K)[9] and good electrical insulation properties[10].

As a comparison, cordierite is another type of filler due to its low thermal expansion, high melting temperature [11], and low dielectric constant [12]. These characteristics are suitable to maintain the dimension of electronic packaging and used for electrical insulation applications. Although cordierite does not provide the high thermal conductivity of hBN, its low thermal expansion and high melting temperature enable it to maintain dimensional stability and enhance mechanical strength under thermal stress. Comparing these fillers independently allows researchers to establish performance baselines for epoxy composites before formulating hybrid systems.

### 3. METHODOLOGY

In this study, EpoxAmite™ 100 epoxy resin supplied by Smooth-On, Inc., USA, with a density of 1.2–1.3 g/mL, was used as the matrix material. The curing agent was diethyl toluene diamine (DETDA) with a purity of at least 97%, obtained from Shandong Aonuo New Material Co. Ltd., China. Hexagonal boron nitride (hBN) powder with an average particle size of 70 nm was supplied by MK Impex Corp., Canada. Cordierite powder with a particle size between 1 and 5 µm was obtained from Standard Advanced Materials, United States. In this study, hBN was chosen for its high thermal conductivity and excellent electrical insulation properties, whereas cordierite was selected for its low thermal expansion, high melting temperature, and mechanical stability under thermal stress.

#### 3.1. Preparation of Cordierite and hBN Composites

The composite was prepared by combining epoxy and hardener, DETDA, with hBN or cordierite loaded at 0, 1, 3, 5, 7, and 9 vol.%, respectively. 0 vol% composition was included as the baseline to evaluate the extent of property improvements due to fillers. The upper limit of 9 vol% was chosen because filler contents beyond this threshold often result in excessively high viscosity, poor dispersion, and agglomeration, which hinder flow into fine gaps and compromise underfill integrity in electronic packaging. hBN or cordierite was mixed mechanically by using a mechanical stirrer for 30 minutes. Then, the composite was ultrasonicated for 15 minutes to ensure uniform dispersion. Ultrasonication of the composite was then done for 15 minutes to make it homogeneous. The curing agent was introduced following the suggested ratio, and it was stirred further for 10 minutes. It was then cast in silicone molds. Curing of the samples was carried out at 100°C in 24 hours and then post-cured at 120 °C for 4 hours.

#### 3.2. Characterisation Techniques

The flexural characteristics of the composites have been determined in the flexure of a three-point bending test on an Instron Universal Testing Machine 5569 as per ASTM D 790. The support span applied was 50 mm, and the crosshead speed was at 2.38 mm/min with dimensions of 60 mm x 12.7 mm x 3 mm. The flexural strength and flexural modulus of the materials were also determined using these conditions. The following equations were used:

$$\text{Flexural Strength, } \sigma_f = \frac{3PL}{2bd^2} \quad (1)$$

where:

- $\sigma_f$  = flexural strength (MPa)
- P = maximum load applied on the specimen (N)
- L = support span length (mm)
- b = specimen width (mm)
- d = specimen thickness (mm)

The same universal testing machine was used to measure the fracture toughness that followed the ASTM D638 procedure by using notched specimens of crack length (4mm). Mechanical tests were carried out in tensile mode, and the loading rate was maintained at 1 mm/min. This has enabled determination of the resistance of the material to crack propagation under stress. Fracture toughness ( $K_{IC}$ ) had been determined by the equation below:

$$\text{Fracture toughness, } K_{IC} = \frac{F_{max}\sqrt{\pi a}}{Bw} f\left(\frac{a}{w}\right) \quad (2)$$

whereby  $f\left(\frac{a}{w}\right)$  is the geometry correction factor, which is given by :

$$f\left(\frac{a}{w}\right) = 1.99 - 0.41\left(\frac{a}{w}\right) + 18.7\left(\frac{a}{w}\right)^2 - 38.48\left(\frac{a}{w}\right)^3 + 53.85f\left(\frac{a}{w}\right)^4$$

A KD2 Pro Thermal Properties Analyzer with an RK-1 sensor was used to measure the thermal conductivity ASTM E1530. The size of the cylindrical specimens was 55 mm in diameter and height, and each specimen had 5 mm holes in the middle to allow making precise and reproducible measurements. The equations used were the following:

$$Q = -KA \left(\frac{\Delta T}{\Delta x}\right) \quad (3)$$

Whereby :

- Q = heat transfer rate (W)
- $\Delta x$  = thickness (m)
- A = area (m<sup>2</sup>)
- $\Delta T$  = temperature difference (K)

The thermal expansion was studied with a Linseis L75 horizontal dilatometer within a temperature range of 30°C to 100°C. A heating rate of 5°C/min was used, and the argon gas was applied as a protective atmosphere. To calculate the coefficient of thermal expansion (CTE), specimens of disc

type measuring 10 mm in diameter and 15-20 mm thick were made. The following equations were used:

$$\epsilon_r = \frac{C X t}{\epsilon_0 X A} \quad (4)$$

whereby:

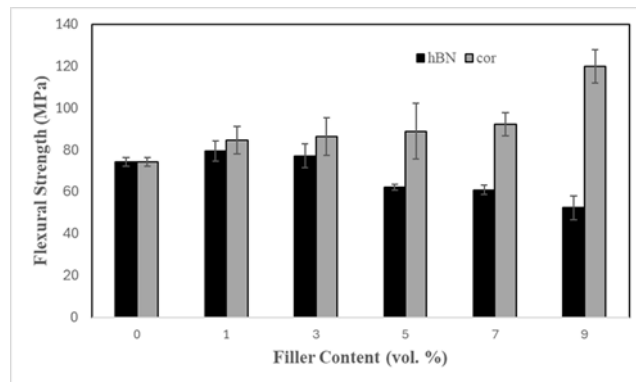
- $\epsilon_r$  = dielectric constant
- $C$  = capacitance in farads
- $\epsilon_0$  = dielectric constant ( $8.854 \times 10^{-12}$  F/m) in a free vacuum
- $A$  is the area of the capacitor plates.

A JEOL JSM-6460LA Scanning Electron Microscope (SEM) was selected to examine the surface morphology and filler dispersion of the composites. The fracture samples were coated with a thin layer of palladium using a JFC-1600 Auto Fine Coater. SEM analysis was evaluated at the accelerating voltages, which ranged from 10 to 15 kV. This test can help to study interfacial bonding, filler agglomeration, and microstructural features of the composites.

## 4. RESULTS AND DISCUSSION

### 4.1. Flexural Strength

The flexural strength of epoxy composites reinforced with two types of fillers, which were cordierite and hBN, was evaluated as shown in Figure 1. The indicated improvement in the strength of cordierite-filled composites was very high, with a maximum strength of 119.90 MPa at 9 vol%. The crystalline structure of cordierite contributes to this improvement by reinforcing the epoxy matrix and enabling more effective stress distribution and load-bearing capacity [13]. In contrast, hBN-filled composites achieved their peak flexural strength at 1 vol.%, which was 79.4 MPa. When the vol.% of hBN increased, the flexural strength of the composites started to decrease. hBN is made up of a layered structure that can help to improve load transfer and ductility [14]. However, at higher concentrations, the particles start to agglomerate. As a result, the uniform stress transfer is disrupted, and the performance is reduced [15]. Both fillers enhance the mechanical properties, though dispersion and optimum loading are very much essential in enhancing maximum strength.

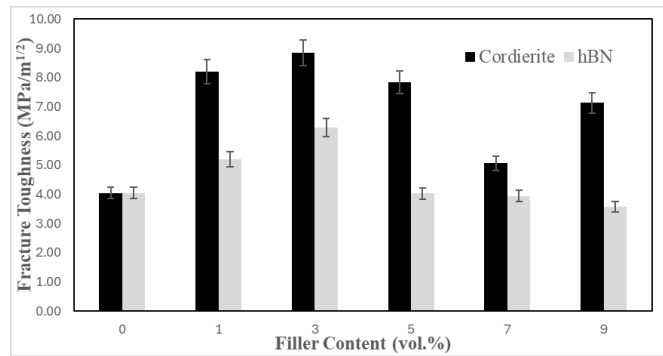


**Figure 1.** Flexural Strength of Epoxy Composites with Different Cordierite and hBN Content.

### 4.2. Fracture Toughness

Figure 2 shows the fracture toughness of epoxy composites reinforced with varying vol.% (0%, 1%, 3%, 5%, 7%, and 9%) of cordierite and hBN, respectively. The neat epoxy recorded a baseline value of toughness of  $4.05 \text{ MPa}\cdot\text{m}^{1/2}$ . By increasing the amount of filler, the fracture toughness of the composites increased because of efficient crack deflection, energy dissipation, and crack bridging around well-dispersed particles [16]. Cordierite showed a better fracture toughness than hBN, especially when the concentration of filler increased. This is because cordierite is a ceramic material that has high strength and thermal stability. These characteristics enhance the load-bearing capacity, facilitate crack deflection, and improve energy dissipation, which contribute to uniform stress distribution

[17]. As the filler contents were increased (5vol.% and 7 vol.%), the fracture toughness of the obtained composites began to reduce because of particle agglomeration. This may break the matrix continuity and form stress concentration points, which reduces the fracture toughness of the composite. However, fracture toughness increased at 9 vol.% because of filler-filler interlocking that enhances structural integrity. In the meantime, at increased loadings (5 vol.%, 7 vol.%, and 9 vol.%), hBN demonstrated a sharp decline in toughness. This is because its plate-like particles form stacks that have weak interfaces with the epoxy matrix. As a result, the crack propagates faster, which reduces the fracture toughness to a level that is lower than that of neat epoxy [18].

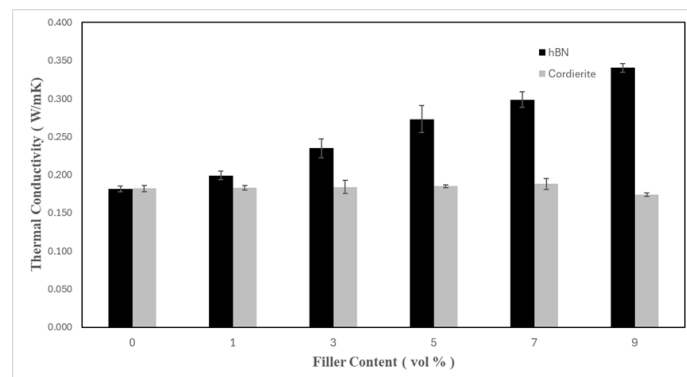


**Figure 2.** Flexural Strength of Epoxy Composites with Different Cordierite and hBN Content.

#### 4.3. Thermal Conductivity

Figure 3 shows the thermal conductivity of epoxy composites reinforced with varying vol.% (0%, 1%, 3%, 5%, 7%, and 9%) of cordierite and hexagonal boron nitride (hBN). The thermal conductivity increased as the filler content of hBN was increased. hBN is well known for its high in-plane thermal conductivity, which can dissipate heat [19]. Increasing the hBN content to 3 vol.% raised the conductivity slightly to 0.235 W/m·K, and further increments to 5 vol.% and 7 vol.% resulted in values of 0.273 W/m·K and 0.299 W/m·K, respectively. The highest conductivity of 0.340 W/m·K was achieved at 9 vol.% of hBN. This trend indicates that the system is achieving the

percolation threshold, which reduces interfacial resistance and allows the photons to transport efficiently via filler-filler contact [20]. Conversely, cordierite has a lower intrinsic thermal conductivity (1.5 to 2 W/m·K). Thus, the addition of cordierite to the epoxy matrix does not improve the thermal conductivity of epoxy composites with a slight increase up to 7 vol.% (0.182 to 0.188 w/m·K). However, thermal conductivity decreased at 7 vol.% loadings. This reduction is related to agglomeration and microstructural defects of filler particles [21]. Overall, hBN is more efficient in increasing thermal conductivity as the concentration of fillers increases.

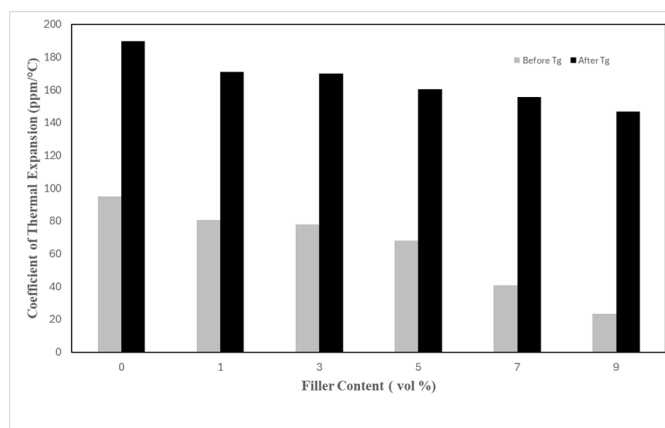


**Figure 3.** Thermal Conductivity of Epoxy Composites with Different Cordierite and hBN Content.

#### 4.4. Thermal Expansion

Figure 4 shows the thermal expansion of epoxy composites reinforced with varying amounts of cordierite at 0 vol.%, 1 vol.%, 3 vol.%, 5 vol.%, 7 vol.%, and 9 vol.% of cordierite before and after the glass transition temperature. Cordierite recorded lower thermal expansion than neat epoxy. The lowest thermal expansion of epoxy composites was recorded at 9 vol.%, which was at 23.44 ppm/°C. This is because cordierite is made up of a crystalline structure, which is very stable under thermal fluctuations [22]. This

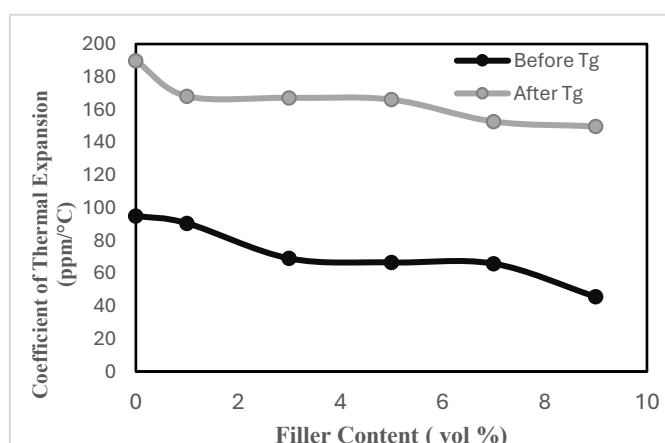
crystal system also helps in improving its mechanical performance when the conditions of temperature are changed [23]. After the Glass transition temperature ( $T_g$ ), the CTE of epoxy composites started to increase because the material changes to a rubbery state and molecular mobility increases [24]. Thus, the filler cannot mitigate thermal expansion effectively. The relatively low CTE values before  $T_g$  demonstrate that cordierite provides adequate dimensional stability during normal device operation, minimising thermal stresses.



**Figure 4.** Thermal Expansion of Epoxy Composites with Different Cordierite Content.

Figure 5 shows the thermal expansion of epoxy composites reinforced with varying amounts of hBN at 0 vol.%, 1 vol.%, 3 vol.%, 5 vol.%, 7 vol.%, and 9 vol.% of hBN before and after the  $T_g$ . The addition of hexagonal boron nitride (hBN) into the epoxy composite helps to decrease the CTE. However, there is an increase in the CTE at 5 vol.% loading of hBN. This occurs because of insufficient mixing or poor surface compatibility with the epoxy resin, which causes agglomeration of particles [25]. The presence of

agglomerates can act as stress concentrators, which affect the thermal properties of the composites [26]. As a result, the CTE value of composites became higher when the temperature increased because of greater dimensional changes. Post- $T_g$  shows a greater CTE value than before  $T_g$  because of the softening of the polymer. However, composites with hBN maintained lower CTE values than pure epoxy, which highlights the stabilising effect of hBN at elevated temperatures [27].

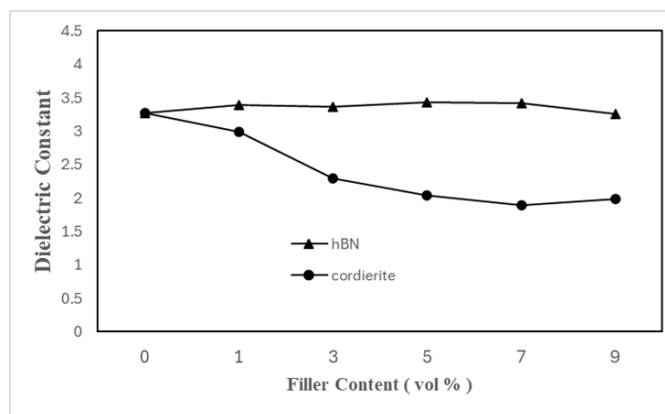


**Figure 5.** Thermal Expansion of Epoxy Composites with Different hBN Content.

#### 4.5. Dielectric Constant

Figure 6 illustrates the dielectric constant of epoxy composites with varying hexagonal boron nitride (hBN) and cordierite content. A pure epoxy recorded a dielectric constant of 3.27. When the hBN content increased, the dielectric constant also increased and peaked at 3.44 with 5 vol.% and decreased to 3.26 at 9 vol.%. This trend occurred because of the high intrinsic dielectric constant of hBN, which enhances polarisation within the composites [28]. However, particle agglomeration may occur and cause the

effective surface area for interaction with the epoxy matrix to be reduced. Thus, the dielectric constant of the composite started to decrease at excessive filler loadings [29]. Contrarily, the dielectric constant of epoxy composites decreased as the cordierite content was increased. The lowest value of the dielectric constant of epoxy composites was recorded at 9 vol.% of cordierite (1.99). This decrease occurs because of the naturally low dielectric properties of cordierite, which translates into reduced polarisation pathways in the composite.

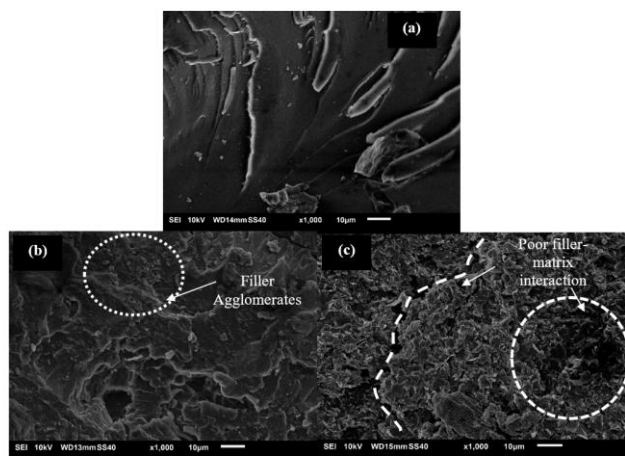


**Figure 6.** Dielectric Constant of Epoxy Composites with Different Hexagonal Boron Nitride and Cordierite Content.

#### 4.6. Surface Morphology

In Figure 7(a), the SEM micrograph of the unfilled epoxy composite shows a smooth and aligned fracture surface. This is typical of brittle behaviour in thermosetting polymers [30]. The results are represented by the homogeneous matrix, which enables the cracks to propagate quickly without being deflected or resisted. The uniform and flat morphology reflects lower energy absorption during failure. There is no mechanism, such as crack pinning or bridging, that will decrease the rate of crack growth. In contrast, Figure 7(b) shows the fracture surface morphology of an epoxy composite filled with cordierite at 9 vol.%. The micrograph depicts the smooth,

aligned morphology as well as plastic deformation along with agglomerations of the fillers. The agglomerates indicate poor dispersion of the filler material. This is because the particles start to cluster together and cannot distribute uniformly throughout the epoxy matrix [31] Next, Figure 7(c) shows the fracture morphology of an epoxy composite filled with hBN at 9 vol.%. The morphology shows a brittle surface due to poor filler dispersion within the epoxy matrix. This composite shows a brittle fracture with crack deflection. This phenomenon indicates poor filler-matrix interaction with more gaps between filler and matrix. The weak bonding between the filler and matrix will cause the cracks to propagate more easily and cause the overall flexural strength of the composites to decrease [32].



**Figure 7.** SEM Micrographs of Epoxy Composites Filled with Single Filler System of (a) Pure epoxy, (b) 9 vol.% of Cordierite, and (c) 9 vol.% of hBN.

#### 5. CONCLUSION

The evaluation of epoxy underfill composites containing a single filler system of hexagonal boron nitride (hBN) and cordierite indicated specific benefits and weaknesses of each of them. Epoxy with cordierite was found to have better mechanical properties with the highest flexural strength (119.90 MPa) and a lower thermal expansion, as the cordierite has low CTE and presented good interfacial bonding. On the other hand, the addition of hBN led to higher thermal conductivity in hBN-filled composites (as high as 0.340 W/m K at 9 vol.%) due to its higher thermal

conductivity properties but lower mechanical strength than cordierite. These findings show that each single filler system improves certain properties but still cannot fulfil all the performance requirements of advanced underfill materials. This scenario highlights the need for combining these two filler systems to become a hybrid filler system to achieve balanced thermal and mechanical characteristics.

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