

Comparison of Experimental and Theoretical CTE of Al/h-BN Metal Matrix Composites

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Abstract

The thermal expansion behavior of aluminum matrix composites reinforced with hexagonal-boron nitride nanoparticles was measured between 30°C and 400°C and compared with theoretical models. The results revealed that the nanoparticle volume fraction had significant effect on the thermal expansion behavior of the composites. The thermal expansion of the composites was within the Turner's and Kerner's models. The experimental thermal expansion of the composites has the closer values to the Schapery's lower bound. The CTE hysteresis was observed negligible between heating and cooling curves due to thermal stability of hexagonal boron nitride within the operational range of temperature.

Keywords: Metal matrix composites, coefficient of thermal expansion, hexagonal boron nitride.

1. INTRODUCTION

Particulate aluminum metal matrix composites have been considered as prospective materials in the applications of electronic packaging and thermal management systems [1, 2]. They have been used in electronic packaging aerospace structures, aircraft and internal combustion engine components. Low thermal expansion and high thermal conductivity are essential for these applications. Most of the studies on metal matrix composites have focused on aluminum as the matrix metal and reinforcements such as: Al₂O₃ [3], SiC [4, 5], B₄C [6], AlN [7], etc. The combination of lightweight, environmental resistance and adequate mechanical properties has made aluminum and its alloys composites very popular.

The present study focuses on determining the effective linear thermal expansion coefficient (CTE) of heterogeneous composite materials. The thermal expansion behavior is particularly important when composite materials are used in conjunction with other materials and when it is necessary to match the CTE of one structural component with another [8-12]. Boron nitride (BN) is heat and chemically resistant refractory compound of boron and nitrogen. It exists in various crystalline forms. The hexagonal boron nitride (h-BN) is the most stable and soft among BN polymorphs. It is used as a lubricant. Because of its excellent thermal and chemical stability, boron nitride ceramics are used as parts of high-temperature equipment.

Table 1: Composition of metal matrix composites

Composite	Composition, vol.%	
	Al	BN
AL-BN-1	90	10
AL-BN-2	85	15
AL-BN-3	80	20
AL-BN-4	75	25
AL-BN-5	70	30

2. MATERIALS AND METHODS

Pure Al powder of 100 μm with 99.9% purity and BN powders of 100 nm were used as the starting materials. Pure powders of Al and BN, in the desired volume fractions, were mixed together by high-energy ball milling for 20 h to ensure the uniform mixing. The mixing was carried out in argon atmosphere to minimize the contamination. The obtained powder mixtures were then sintered to bulk specimens by hot pressing at 800 $^{\circ}\text{C}$ with a pressure of 50 MPa in vacuum, followed by quickly cooling to room temperature in 30 min. In this study, five different composites were prepared (Table 1).



Figure 1: Preparation of Al/h-BN specimens.

The thermal expansion was then measured with a dilatometer (DIL 802) between 100 and 300 $^{\circ}\text{C}$ at heating and cooling rates of 5 $^{\circ}\text{C}/\text{min}$ in argon. With this instrument the difference in length between the specimen to be investigated and a reference sample is

measured, which results in a resolution of $\pm 0.01 \mu\text{m}$. Specimens with a diameter of 5 mm and length of 10 mm were used for the measurement of CTE. The instantaneous CTE at a given temperature was calculated using the following equation:

$$\text{CTE} = \frac{\partial}{\partial T} \left(\frac{\Delta L}{L} \right) \quad (1)$$

where L is the length of the specimen and T the temperature. All of the specimens were tested from 30°C to 400°C for heating cycle and 400°C to 30°C during cooling cycle at 10°C/min. this temperature range was selected so as to include the entire usable range of the composite, without the formation of liquid phase in the matrix.

3. RESULTS AND DISCUSSION

Due to high pressure employed during fabrication, the composites were free of porosity and microscopically homogeneous as shown in figure 2.

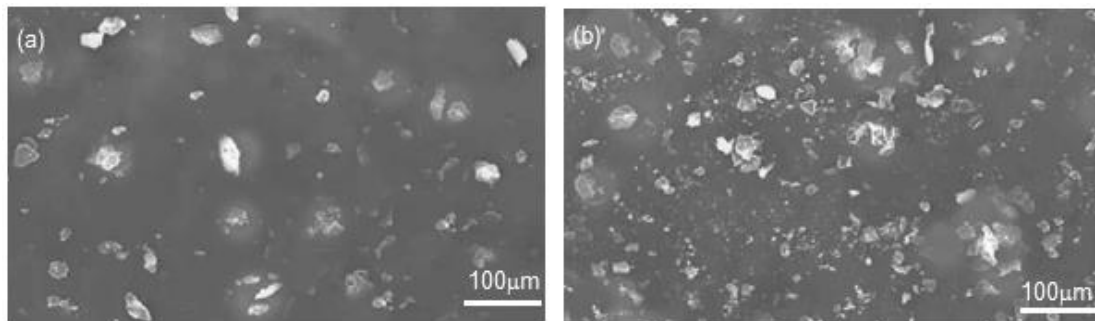


Figure 2: Particle distribution in Al matrix: (a) 10% h-BN and (b) 30% h-BN.

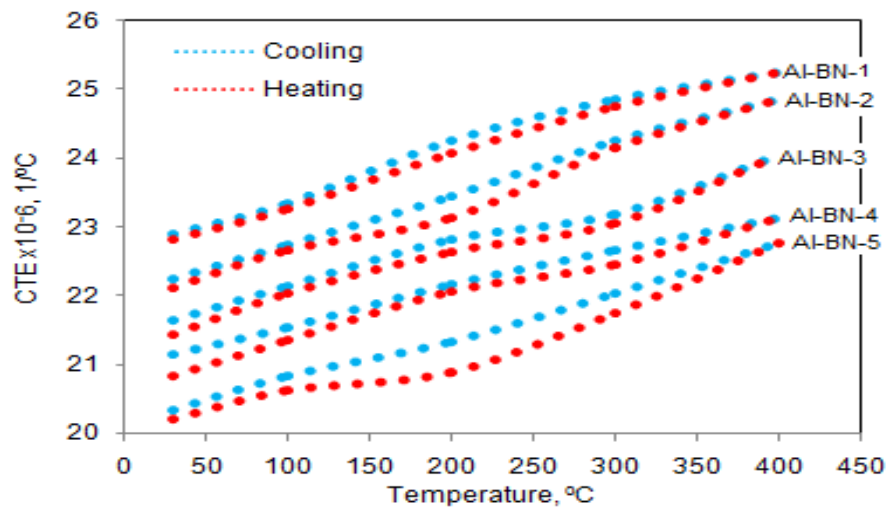


Figure 3: Coefficient of thermal expansion as a function of temperature for: (a) AL-BN-1, (b) AL-BN-2, (c) AL-BN-3, (d) AL-BN-4 and (e) AL-BN-5.

3.1 Coefficient of thermal expansion

The CTE results with the variation of temperature for the composites are shown in figure 3. The CTE measured during the heating cycle increases with increasing temperature between 30 and 400°C for all the composites. The CTE measured during the cooling cycle decreases continuously with decreasing temperature in the same trend but above of the heating cycle. The hysteresis of CTE was negligible due its thermal stability within the operating range 30 to 400°C. The Al/BN composite is a two-phase system composed of a continuous Al matrix and isolated BN nanoparticles each having different mechanical and thermal properties. So the thermal expansion behavior of the composite is the result of the interaction between Al matrix and BN nanoparticles through interfaces. The small difference in CTE during heating and cooling is on account of CTE mismatch between Al and h-BN.

3.2 Comparison of CTEs among experimental and theoretical results

In order to comprehend the thermal expansion behavior of the composites well, it is imperative to judge against quantitatively theoretical predictions with experimental results. Many analytical and semi-empirical formulas have been derived to predict the linear thermal expansion of heterogeneous composites [13-15]. The rule-of-mixture models are derived from the assumption of uniform strain or stress of the composite structure.

$$\alpha_c = \alpha_r V_r + \alpha_m V_m \quad (2)$$

Turner model [13] takes into account the mechanical interaction between the phases in the heterogeneous composite material.

$$\alpha_c = \frac{\alpha_m V_m K_m + \alpha_r V_r K_r}{V_m K_m + V_r K_r} \quad (3)$$

Another model for particulate composites is given by Kerner [14], which accounts for both shear and isostatic stresses developed in the component phases, and gives the CTE for the composite as:

$$\alpha_c = \alpha_m V_m + \alpha_r V_r + V_m V_r (\alpha_r - \alpha_m) \frac{K_r - K_m}{V_m K_m + V_r K_r + 3K_m K_r / 4G_m} \quad (4)$$

Schapery [15] used elastic energy principles to derive bounds for effective CTEs of anisotropic composites made from isotropic constituents. The lower and upper bounds for the effective coefficients are given by

$$\alpha_c^u = \alpha_m V_m + \alpha_r V_r + \frac{4G_m (K_c - K_r)(\alpha_m - \alpha_r)V_r}{K_c (4G_m + 3K_r)} \quad (5)$$

$$\alpha_c^l = \alpha_m V_m + \alpha_r V_r + \frac{4G_r (K_c - K_m)(\alpha_r - \alpha_m)V_m}{K_c (4G_m + 3K_m)} \quad (6)$$

Where α is the CTE, v is the volume fraction, K is the bulk modulus, G is the shear modulus, and subscripts c , m , r refer to the composite, matrix and particle respectively.

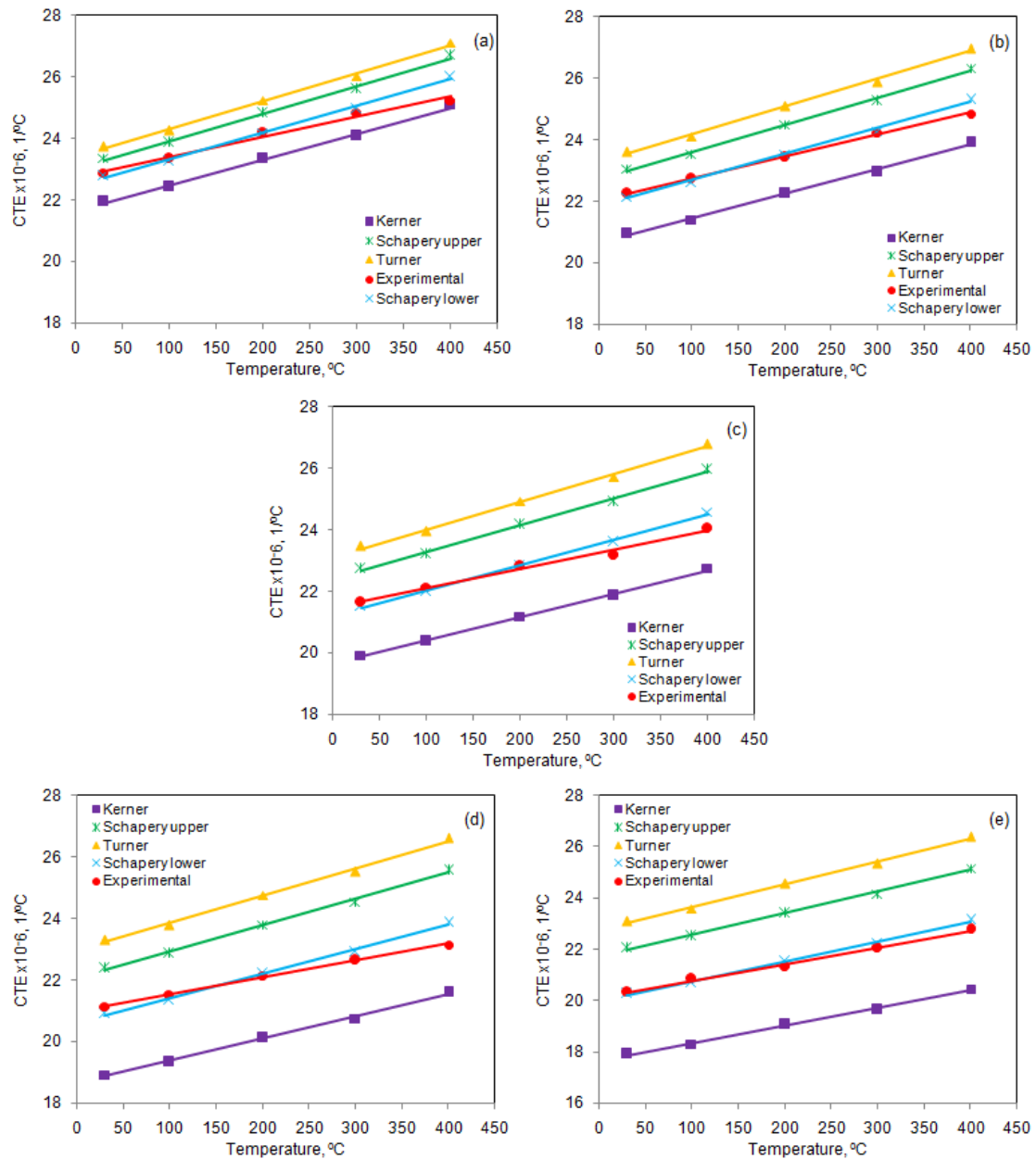


Figure 4: Coefficient of thermal expansion as a function of temperature compared with Schapery and Turner models for: (a) AL-BN-1, (b) AL-BN-2, (c) AL-BN-3, (d) AL-BN-4 and (e) Al-BN-5.

Figure 4 compares the experimental results with the theoretical models for all five composites. It is worth noting that the upper bound of composite CTE is Turner’s model and the lower bound of composite CTE is Kerner’s model. At low temperature, the experimental CTEs agree relatively with the values predicted by Schapery’s lower bound; while the CTEs at higher temperature seem to be lower than the values estimated by Schapery’s lower bound. The experimental CTEs for entire temperature

range show significantly deviation from Turner's model. This is owing to Turner's model is based on the fact that only uniform hydrostatic stresses exist in the phases, while the stresses inside the composite are very complex. Kerner's model, which assumes spherical particles, may underestimate the actual constraints in the composite.

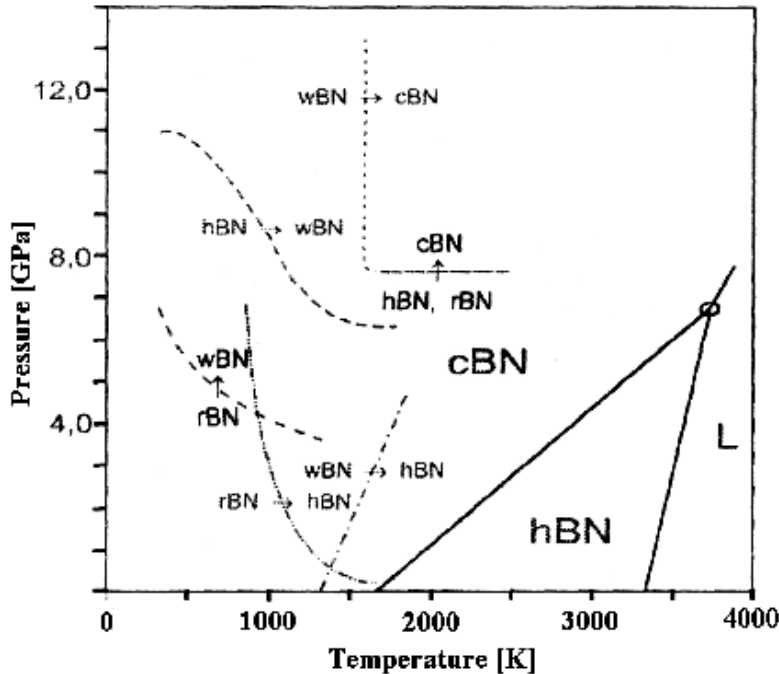


Figure 5: The P-T phase equilibrium diagram of boron nitride and the kinetic lines for the direct transformations between the BN polymorphs.

The phenomena behind the thermal stability of h-BN below the melting point of Al (660.32°C) is due to its stability to decompose at temperatures at 1000 °C in air, 1400 °C in vacuum, and 2800 °C in an inert atmosphere. The phase equilibrium diagram depicted in figure 5 reproduced from reference [16] shows the phase transformations between two BN polymorphs. Among the transformations indicated in figure 5 the most interesting one from a practical standpoint is the h-BN → c-BN transformation. Figure 6 depicts the crystal structures of h-BN and c-BN. A common feature is the location of B and N atoms as nearest neighbors in alternate positions. The crystal lattice of h-BN consists of the stacking sequence of the hexagonal layers. The strong σ covalent bond between B and N atoms within the hexagonal layers has an ionic component due to the presence of two atomic species with different electronegativity. The interatomic bond between two successive hexagonal layers is extremely weak. The crystal lattice of c-BN is cubic. The c-BN has tridimensional lattices in which each atom is in a sp^3 hybridization state and establishes four strong covalent σ bonds with its nearest neighbors that make up a tetrahedron around it. The cubic form (c-BN) is less stable than the hexagonal form (h-BN), but the conversion rate between the two is negligible at room temperature. The partly ionic structure of

BN layers in h-BN reduces thermal conductivity, whereas the interlayer interaction increases hardness.

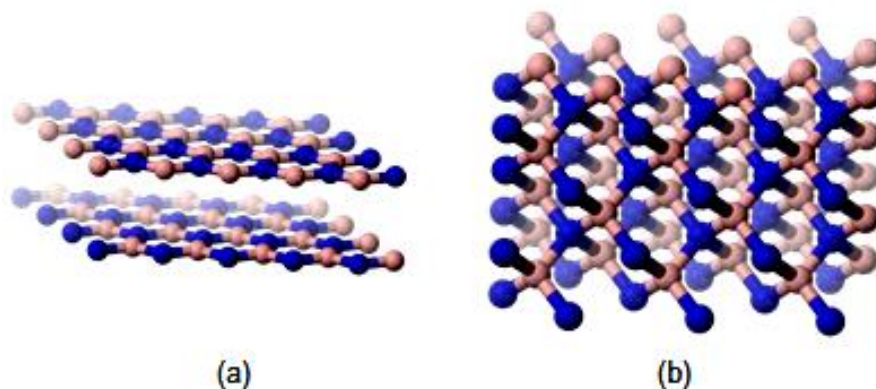


Figure 6: Crystal structures of BN: (a) h-BN and (b) c-BN.

Therefore, the present work suggests that Al matrix composites reinforced with h-BN are suitable for structural applications wherein excellent thermal stability and low thermal expansion are of high priority.

CONCLUSIONS

In this research, the thermal expansion behavior of Al-based composites reinforced with hexagonal-boron nitride nanoparticles has been investigated. The results indicate that the volume fraction of nanoparticle can have significant effect on thermal expansion behavior of the composites. At lower temperature, the CTEs were within the Schapery's bounds, while the CTEs at high temperature were near to the values predicted by Kerner's model.

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