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Thermal Expansion and Thermal Conductivity Behaviors of Al-Si/SiC/graphite Hybrid Metal Matrix Composites (MMCs)

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Aluminum-silicon based hybrid composites reinforced with silicon carbide and graphite particles were prepared by liquid phase particle mixing (melt stirring) and squeeze casting. The thermal expansion and thermal conductivity behaviors of hybrid composites with various graphite contents (5.0; 7.5; 10 wt.%) and different silicon carbide particle sizes (45 µm and 53 µm) were investigated. Results indicated that increasing the graphite content improved the dimensional stability, and there was no obvious variation between the thermal expansion behaviors of the 45 µm and the 53 µm silicon carbide reinforced composites. The thermal conductivity of hybrid composites was reduced due to the enrichment of the graphite component.

Keywords: hybrid MMC, thermal expansion, thermal conductivity.

1. INTRODUCTION

Metal matrix composites (MMCs) can withstand the extreme conditions often encountered in automotive and space environments [1]. In particular, particle reinforced metal matrix composites (PMMCs) possess distinct advantages over fiber reinforced counterparts in terms of low cost, isotropic mechanical properties and the ability to be processed using technology similar to that used for monolithic materials [2–4]. Aluminum MMCs reinforced with silicon carbide (SiC) particles have up to 20 % improvement in yield strength, a lower coefficient of thermal expansion and a higher modulus of elasticity, and they are more wear resistant than the corresponding non-reinforced matrix alloy systems [5]. By varying the matrix, reinforcement and volume fractions, the MMCs can be customized to provide a good coefficient of thermal expansion (CTE) matching for thermal management and thermal conductivity (TC) applications [6]. It is essential to evaluate new materials for their thermal stability and to measure their properties including CTE and TC for specialty products, such as break discs made from castings, before actual use. It is expected Al-Si/SiC/graphite hybrid composites can be used as load bearing material for such kind of applications. In the open literature there is no comprehensive work for Al-Si/SiC/graphite hybrid composites to determine thermal properties.

In this work, the thermal expansion and TC of an aluminum-silicon based hybrid MMC reinforced with SiC and graphite was investigated in terms of the effect of their content and the particle size.

2. EXPERIMENTAL PROCEDURES

During the production of the hybrid composite, the matrix alloy (Al-11.8 wt.% Si) was melted in a furnace with a crucible capacity of about 10 kg. The furnace heated up to 700 °C for 30 min; then, the liquid was stirred with an Al_2O_3 plasma sprayed steel impeller to avoid aluminum-iron interactions. SiC particles with an average size of 45 µm and 53 µm and graphite particles with an average size of 90 µm were introduced into the formed vortex simultaneously. Graphite particles were polycrystalline and irregular in shape which has a purity of 99.8 % and 2.2 g/cm^3 density. The amount of graphite particles was varied (5.0; 7.5; 10 wt.%) for possible bearing applications for each particular SiC particle size addition with a consistent reinforcement fraction (~20 vol.%). For the purpose of improving surface wettability and interface bonding between matrix alloy and graphite particles, the surface of graphite particles was previously coated with copper by a cementation process using aqueous and supersaturated copper sulphate solution. Details of the process can be found elsewhere [7–8].

Mixing was constantly applied until the composite melt attained a mushy state. Stirring was continued for 5 min to increase the mechanical locking of the SiC and graphite particles into the aluminum matrix. After the mixing was completed, the temperature was allowed to reach 700 °C, and the mixture was stirred for an additional 20 s to prevent settling of the SiC and graphite particles. Then, it was poured in to a squeeze casting steel mould, which was preheated to 500 °C. The solidification was completed under 50 MPa. The melting, introduction of the reinforcement particles and mixing were all conducted under a protective argon atmosphere to avoid the oxidation of both the melt and reinforcement particles.

All thermal expansion and CTE measurements for prepared specimens (20 mm × 5 mm × 5 mm) were conducted in the research grade dilatometer system (Anter-Unitherm Model-1161V). All the experimental runs were conducted in the temperature range of 20 °C to 400 °C under the protective argon gas atmosphere. The heating and cooling rates for measurements were 5 °C/min and were

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continuously monitored by the computer based data acquisition system. Thermal conductivity was measured by the laser flash method with a thermal conductivity meter (Netzsch LFA-447) in the temperature range of 50 °C to 300 °C. Disc specimens with dimensions of 25 mm in diameter and 2 mm in thickness were cut from the squeeze cast composites for the thermal conductivity testing.

Microstructural characterization studies were performed by using optical microscopy and scanning electron microscopy (SEM, Jeol-JSM 6060LV) equipped with energy dispersive spectroscopy (EDS).

3. RESULTS AND DISCUSSION

3.1. Microstructures of composites

The microstructures of hybrid composites reinforced with SiC and graphite particles are shown in optical microscopy images (Fig. 1). For brevity, the microstructures of hybrid composites for a consistent reinforcement of graphite particles (10 wt.%) for each particular SiC particle size addition (45 μm and 53 μm) are presented in both optical and SEM images. The distribution of SiC and graphite particles is random with no cracks and deleterious pores in the microstructure. The measured microscopic porosities were found between 2 % – 3.5 % for produced hybrid composites.

Interdendritic segregation is observed because the particles were pushed out by the solidification front and they are preferentially located in the eutectic regions during solidification. The presence of the particles in the matrix particles considerably refines the microstructure, impeding the coarsening of the dendrites of the primary phase during solidification.

The SiC particles moved mostly at the primary aluminum dendrite boundaries, although some are observed within the aluminum grains. Porosity, which was revealed after slight etching of the specimens, could often be observed in the region of SiC particle clusters. Experimental observations showed that introducing graphite particles revealed similar effect with SiC addition. Increasing graphite content in the composite matrix leads grain refinement for both primary aluminum dendrites and eutectic silicon. The microstructural investigation also showed that silicon was present around the SiC particles and was located on the SiC surfaces (Fig. 1, a and c). Some of the primary silicon crystals were also found adjacent to SiC particles. Some of the primary silicon particles were heterogeneously nucleated on the SiC particles (Fig. 1, b) and equiaxed silicon particles can be seen around the large SiC dispersoid (Fig. 1, d).

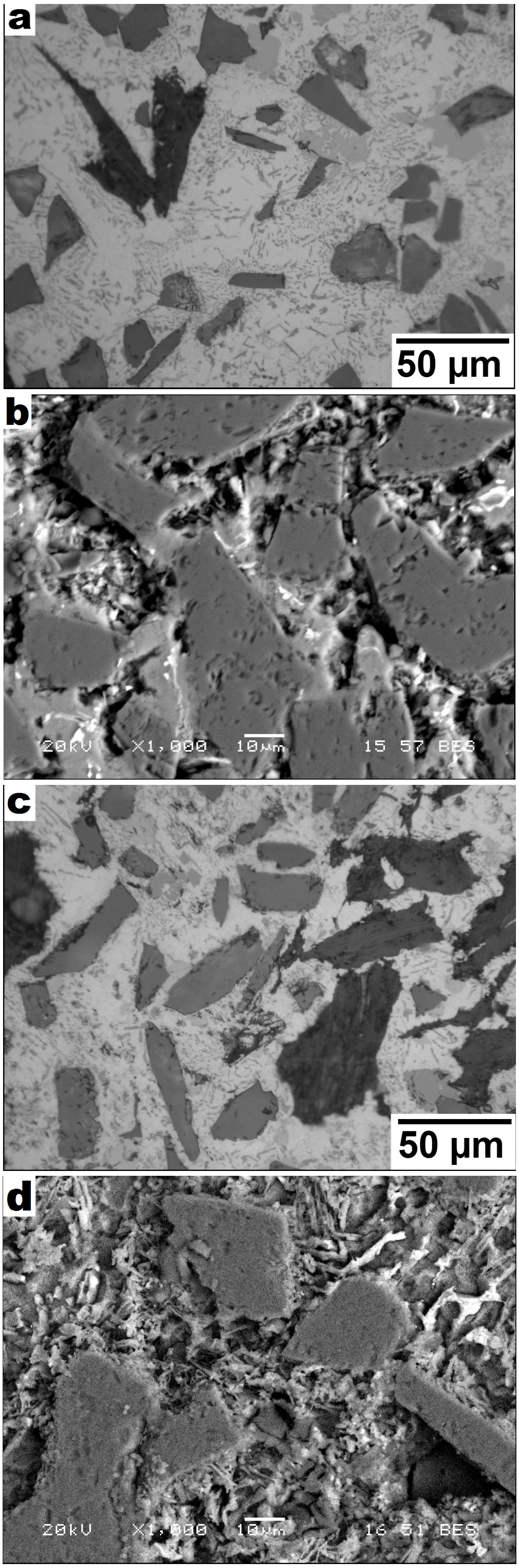

Fig. 1. Optical and SEM microstructures of some selected Al-SiC-graphite hybrid composites: a, b – 20 vol.% SiC (45 μm) / 5.0 wt.% graphite; c, d – 20 vol.% SiC (53 μm) / 10 wt.% graphite

3.2. Thermal expansion behaviors of composites

The relative thermal expansion behavior of aluminum-silicon based hybrid MMCs reinforced with SiC and graphite particles measured at temperatures varying from 20 °C to 400 °C for a single thermal cycle are shown in Figs. 2 and 3. Since there is no evident variation in heating and cooling curves, figures regarding 5.0 wt.% graphite content not presented for the sake of clarity.

Thermal strain can be attributed to thermal stress. Higher thermal stress can lead to the generation of strain

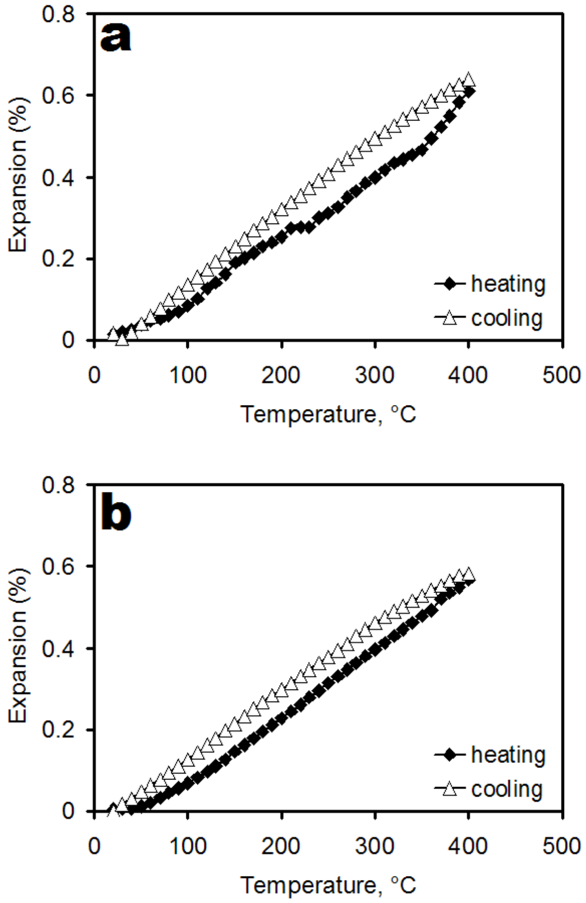

Fig. 2. Thermal strain response curves recorded during the heating and cooling between 20 °C and 400 °C for the hybrid Al-Si-20 vol.% SiC (53 μm)/graphite composites, 7.5 wt.% graphite (a) and 10 wt.% graphite (b)

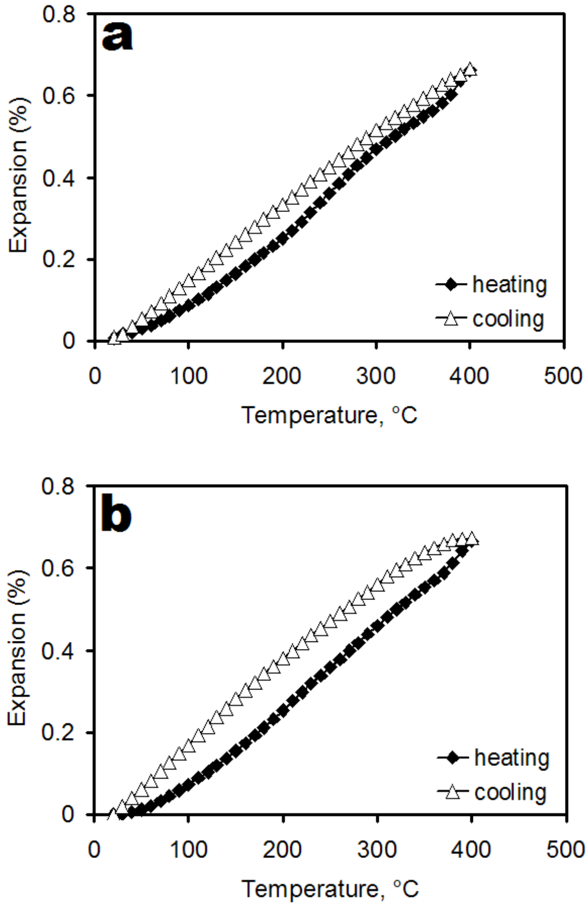

Fig. 3. Thermal strain response curves recorded during the heating and cooling between 20 °C and 400 °C for the hybrid Al-Si-20 vol.% SiC (45 μm)/graphite composites, 7.5 wt.% graphite (a) and 10 wt.% graphite (b)

hysteresis between the heating and cooling cycles and to the retention of residual strain as the result of the plastic deformation or yielding of materials. Thermal response curves can provide valuable information for predicting the thermal stability, failure/damage and life of the structural materials that have been subjected to heating and cooling conditions [9, 10].

Figs. 2 and 3 show that hysteresis loops of investigated hybrid MMCs were induced with increasing amount of graphite for both (45 μm and 53 μm) particle sizes of SiC reinforcements. The results show that the thermal strain of all hybrid composites increased as the amount of graphite increased, indicating that introducing a high amount of graphite to the Al-Si based MMC was not very beneficial to the dimensional stability. The effect of SiC particle size on the thermal strain of this Al-Si based hybrid MMCs showed no obvious pattern.

The results of CTE measurements in the temperature range of 20 °C to 400 °C are shown in Fig. 4, a and 4, b. The CTE of the Al-Si based hybrid MMCs is obviously lower when the graphite content is increased for both (45 μm and 53 μm) particle sizes of the SiC reinforcements. However, only the hybrid composite reinforced with 7.5 wt.% graphite showed sharp CTE decrease beyond 250 °C. Figs. 4, a and 4, b also show that the CTE of the composites increased with the temperature, reached a saturation with further increase of the temperature beyond 200 °C – 250 °C; depending on the graphite content. The higher the graphite content the lower the saturation temperature.

Measurements in the temperature range of 20 °C to 400 °C revealed a marginal reduction in the average CTE of the composites with the addition of graphite particles as reinforcement. The maximum CTE value obtained for the 5 wt.% graphite and 53 μm SiC reinforced Al-Si based hybrid MMC was recorded at 330°C.

Increasing the graphite content to 7.5 wt.% and 10 wt.% resulted in a shift of this maximum temperature for CTE value to 240 °C and 180 °C, respectively. The tendency was similar for both 45 μm and the 53 μm SiC reinforced composites: a decrease of the maximum CTE value with increased temperature.

The decrease of the maximum temperature for CTE values for graphite reinforced composites is considered as a result of the relaxation of the compressive stress in the matrix, which was also stated by Fei et al. [11], who studied Al/AlBO$_w$ containing Fe$_3$O$_4$ particle composites produced by the squeeze casting method. The reduction in CTE values can also be attributed to the lower CTE value of graphite compared to the Al-Si matrix alloy and SiC reinforcement and the ability of the reinforcements to effectively constraint the expansion of the matrix. It is reported that, SiC and graphite has a CTE of about 4.5 10^{-6}/°C and 4.06 10^{-6}/°C in the temperature range of

20 °C – 400 °C, while the compared value of Al-12% Si alloy about 22.3 10^{-6}/°C in the temperature range of 50 °C – 300 °C, respectively [12, 13, 9].

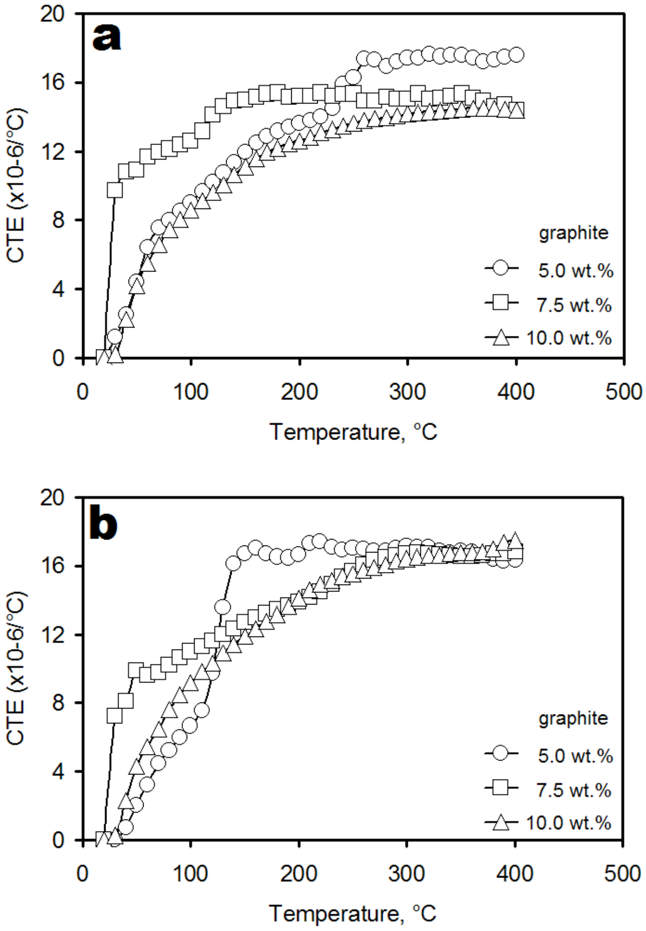

Fig. 4. Coefficient of thermal expansion as a function of temperature for the Al-Si-20 vol.% SiC (53 µm)/graphite composites (a), and Al-Si-20 vol.% SiC (45 µm)/graphite composites (b)

The CTE of particle reinforced MMCs is affected by a variety of factors, such as interfacial reactions, plasticity due to CTE mismatch between particle and matrix during heating or cooling, and residual stresses [14]. As can be seen from the hysteresis between the heating and cooling curves, increasing the amount of graphite reinforcement in SiC reinforced Al-Si based hybrid MMCs indicates higher residual thermal strains (Figs. 2 and 3) but lower CTE values.

The incorporation of ceramic reinforcements in metallic matrices of MMCs generates residual stresses during cooling from the material processing temperature due to the large difference between the CTEs of the reinforcement and matrix [2]. Residual stresses cause compressive stresses on the reinforcements and tensile stress on the matrix, and their magnitude varies with the characteristics of reinforcement and matrix as well as with the processing [9, 10, 15]. Ren et al. [10] stated that such a tensile stress is considered to be generated from the CTE mismatch between the matrix and reinforcements, progressively diminished approaching to zero during the heating stage. Therefore, the compressive stress in the matrix initiated to build-up with the temperature since the expansion of matrix is constrained by the vicinity of reinforcement particles. Accordingly, compressive stress begins to accumulate to such an intensity until it surpasses the yield strength of the matrix which lessening with the temperature elevation, ensuing the plastic relaxation. The tensile stress builds-up throughout the reduction in temperature during subsequent cooling period and new residual stresses will be generated. However, the matrix has deformed and work-hardened enough to result in less noticeable plastic relaxation of the matrix, arising from the new tensile stress, increasing with the reduction in temperature [2, 6, 9]. Introducing the third phase (graphite) to the SiC reinforced Al-Si composite induced residual strain in the Al matrix. Due to the thermal expansion mismatch between graphite and the metal phase, residual stresses are expected to be tensile in the metal phase and compressive in the graphite, and, during heating, the residual stresses relaxed elastically or plastically [16]. This relaxation process was observed in this investigation with an increase in graphite content, resulting in an open hysteresis during the heating and cooling cycle.

3.3. Thermal conductivity of composites

Table 1 shows the thermal conductivity results obtained at different temperatures for SiC reinforced Al-Si based hybrid MMCs with various graphite contents. TC decreases with increasing graphite content. As the amount of graphite increases from 5 wt.% to 7.5 wt.%, there is a slight decrease in TC from 202.2 W/mK to 194.8 W/mK for the 53 µm SiC reinforced Al-Si based hybrid MMC at a 50 °C testing temperature. With a further increase of graphite content to 10 wt.%, TC then decreases to 185.6 W/mK. Increasing the test temperature also resulted

Table 1. Thermal conductivity values (W/mK) of the Al-Si hybrid composites between 50 °C and 300 °C

Composite	Test temperature, °C					
	50	100	150	200	250	300
Al-Si 20 vol.% SiC (45 µm)/5.0 wt.% graphite	186.4	178.8	171.1	165.4	161.2	158.1
Al-Si 20 vol.% SiC (45 µm)/7.5 wt.% graphite	181.2	173.5	157.6	148.2	138.8	130.5
Al-Si 20 vol.% SiC (45 µm)/10 wt.% graphite	176.4	165.5	152.4	136.3	128.8	118.4
Al-Si 20 vol.% SiC (53 µm)/5.0 wt.% graphite	202.2	195.5	188.4	171.5	168.3	158.6
Al-Si 20 vol.% SiC (53 µm)/7.5 wt.% graphite	194.8	190.4	181.1	170.5	162.1	150.8
Al-Si 20 vol.% SiC (53 µm)/10 wt.% graphite	185.6	175.5	166.2	155.3	146.2	136.7

in a continuous decrease in TC values. The TC value for 5 wt.% graphite and 53 μm SiC reinforced MMC showed a value of 195.5 W/mK at 100 °C and continuously decreased with increasing test temperature, falling to 158.6 W/mK at 300 °C. The 45 μm SiC reinforced hybrid MMCs showed a similar trend in terms of TC variation as a function of graphite content and temperature. However, the TCs of the 45 μm SiC reinforced hybrid MMCs were lower than those of the 53 μm particle size SiC reinforced composites, as shown in Table 1.

The dependence of the overall conductivity on the particle diameter for spherical particles of equal size was investigated with several predictions [17 – 19]. The reason for the decrease of the TC values with decreasing grain size of SiC can be attributed to the interfacial properties between the Al matrix and SiC particles. It is obvious that decreasing the grain size results in a larger surface area between the Al matrix and SiC particles. It is also possible to form a greater amount of interfacial reactions by increasing the interfacial surface area. It was stated by Ren et al. [10] that the interfacial reaction between SiC particles and Al matrix can reduce the TC of the composites. Ruch et al. [20] also pointed out that decreasing the interfacial bond strength in Al-Si-diamond MMCs resulted in decreased thermal conductivity. Although it was stated that [21] porosity can severely degrade the thermal and mechanical properties of the MMCs, the SiC and graphite particles were distributed uniformly in the aluminum matrix (as previously shown in Figs. 1), and no considerable level of pores were observed in the present study when the amount of graphite reinforcement reached 10 wt%.

Because of the lower TC of graphite itself (about 150 W/mK) compared with that of Al (237 W/mK), Si (150 W/mK) and SiC (180 W/mK) [10], the TC of aluminum matrix decreased with the increase of the graphite content so that the TC of the composites exhibited a similar tendency of deviation. It can be concluded that the composites with 10 wt.% graphite content would exhibit low TC compared with those of the composites with 5 wt.% to 7.5 wt.% for a consistent level of porosity.

4. CONCLUSIONS

The following major conclusions can be drawn from this investigation.

1. The distributions of SiC and graphite particles in the MMCs were homogeneous and macroscopically free of pores indicating the production of the hybrid composites were succeeded.
2. Introducing graphite particles into Al-Si based hybrid MMCs reinforced with SiC particles (~20 vol.%) resulted in decreased thermal expansion of the composites, since SiC has lower thermal expansion coefficient than that of aluminum matrix.
3. Introducing a high amount of graphite to the Al-Si matrix alloy was found to be beneficial to the dimensional stability of SiC reinforced Al-Si based hybrid MMCs. Results revealed that graphite particles absorb the thermal expansion because of their layered structure.
4. The CTE of the Al-Si based hybrid MMCs was found to be lower when the graphite content was increased for both (45 μm and 53 μm) SiC reinforcements of these composites.
5. Increasing the graphite content for both (45 μm and 53 μm) SiC reinforced Al-Si based hybrid MMCs resulted in a shift of the maximum temperature of the CTEs, because of the relaxation of compressive stresses in the matrix.
6. TC was found to decrease as the content of reinforcement and the temperature increases, since the reinforcements have lower TC values than that of matrix alloy and because, increased temperature diminish the thermal diffusivity.

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