Thermal Expansion Behavior of SiCP/Aluminum Alloy Composites Fabricated by a Low-Pressure Infiltration Process*1

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Thermal cycling test was carried out for 20 vol% and 40 vol% SiC particle/Al-4 mass%Cu alloy composites to evaluate the bonding strength of the SiC particle/matrix interface in PRMMC fabricated by a low-pressure infiltration process (LPI process). The SiC particles were distributed homogeneously in the specimens and a reaction layer of less than 1 μm thickness was observed at the SiC particle/matrix interface before thermal cycling test. This reaction layer was identified as Al4C3 formed by the reaction between the alloy melt and SiC particle during infiltration. The 9 phase, which might form a coherent interface with Al4C3, crystallized around the SiC particles through the Al4C3 layer. The coefficient of thermal expansion was hardly changed during thermal cycling for both the 20 vol% and 40 vol% SiC particle/Al-Cu composites. No significant change in the microstructure and no detachment at the SiC particle/matrix interface were observed after thermal cycling test. The interfacial structure consisting of SiC, Al4C3, 9 phase and α-Al in order was considered to exhibit a strong bonding of SiC particles to the matrix. The Vickers microhardness measured on the SiC particles in the specimens having a strongly bound interface show hardness values with a small scatter, while those for the specimens having a weakly bound interface exhibit a large scatter in the hardness values. It is suggested that the bonding strength of the reinforcement particle/matrix interface could be evaluated qualitatively from the Vickers microhardness test.

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1. Introduction

Metal matrix composites (MMC) are widely studied as prime candidates for the structural materials for transportation equipments such as spacecraft, aircraft and automobile. Among the fabrication processes for MMC, the casting process is superior because of its high productivity. To obtain sound castings, a high pressure of several hundreds MPa is required in the conventional infiltration processes and the so-called high pressure infiltration process is widely adopted. However, the products are expensive since a large-scale facility for a high pressure infiltration is indispensable. On the other hand, the low-pressure infiltration process (LPI process), developed by the present authors, can fabricate the particle reinforced MMC (PRMMC) under an infiltration pressure of about 0.2 MPa by using the mixed powder consisting of reinforcement particles and metal particles1,2) to control the volume fraction and distribution of the reinforcement particles accurately.

In the present work, to evaluate the bonding strength of the interface between the reinforcement particle and the matrix in PRMMC fabricated by LPI process, the thermal expansion behavior was investigated by thermal cycling test.

2. Experimental

Al-4 mass%Cu alloy was used as matrix and SiC particle was used as reinforcement. The reinforcement particles were screened with sieves and the particle size was distributed in the range from 50 to 100 μm. The mixed powder consisting of reinforcement particles and pure aluminum particles (particle size: 100 μm) in various volume fraction was prepared to control the volume fraction of reinforcement particles in PRMMC. The low-pressure infiltration apparatus is shown in Fig. 1. Silica tube of 15 mm in inner diameter was used as a container that had a small nozzle at the bottom to release the air inside the silica tube. Coarse alumina balls (particle size: 1 mm) were laid at the bottom of the silica tube as a bed layer. The mixed powder and the Al-4 mass%Cu alloy were piled up in order in the silica tube as shown in Fig. 1. The upper Al-4 mass%Cu alloy was melted by high frequency induction heating and the melt temperature was held at 1123 K, then the alloy melt was forced to infiltrate into the mixed powder layer by applying an argon gas pressure of 0.2 MPa on the melt surface. Pressing was stopped after some extent of the alloy melt flowed out through the nozzle. The dimension of PRMMC specimens obtained was 15 mm in diameter and 20 mm long. The microstructure and solute distribution in the PRMMC specimens were examined on the horizontal section with optical microscope, EDX-SEM and TEM. Thermal cycling test was carried out from room temperature to 473 K.
using TDA in order to evaluate the thermal expansion behavior and the bonding strength of the interface between reinforcement particle and matrix. After thermal cycling test, the microstructures of the PRMMC specimens were examined on the horizontal section with EDX-SEM and TEM and were compared with the microstructures before thermal cycling test. To evaluate the bonding strength of the interface between reinforcement particle and matrix, Vickers microhardness was measured on the reinforcement particles in the PRMMC specimens, using AKASHI MVK-H1 hardness tester with 0.098N load.

3. Results and Discussion

3.1 Thermal expansion behavior of PRMMC

Figure 2 shows the results of thermal cycling test carried out on the 20% vol% and 40 vol% SiC particle/Al-Cu composites. The coefficient of thermal expansion was hardly changed. It has been reported that the serration and extended displacement in the load-displacement curves obtained by the three-point bending tests might indicate the detachment occurring at the interface between matrix and reinforcement. In the present work, no serration was observed in Fig. 2, indicating that no detachment occurred at the interface during thermal cycling test in the both specimens. The average coefficients of thermal expansion of the composites in the temperature range between R.T. and 473 K are shown in Fig. 3 together with the theoretical ones calculated based on the rule of mixtures, Schapery’s model and Turner’s model. Shapery derived the effective coefficient of thermal expansion based on Shapery’s model is given by eq. (1). The upper and lower bounds of $K_C$ (Hashin’s bounds) is given by eqs. (2) and (3). On the other hand, considering the constraint due to a dimensional change of each component, the coefficient of thermal expansion derived by Turner is given by eq. (4);

$$\alpha_C = \alpha_P + (\alpha_M - \alpha_P) \left( \frac{1}{K_C} - \frac{1}{K_P} \right) \left( \frac{1}{K_M} - \frac{1}{K_P} \right)$$

$$K_{C,upp} = K_M + \frac{1}{K_M - K_P} \left( 1 - V_P \right) \left( 1 - V_P \right) \left( K_M + 4G_M/3 \right)$$

$$K_{C,low} = K_P + \frac{1}{K_P - K_M} \left( 1 - V_P \right) \left( 1 - V_P \right) \left( K_P - 4G_P/3 \right)$$

$$\alpha_C = \frac{V_P \alpha_P K_P + (1 - V_P) \alpha_M K_M}{V_P K_P + (1 - V_P) K_M}$$

where $\alpha$ is the coefficient of thermal expansion, $V$ is the volume fraction and $K$ is the bulk modulus, and the subscripts C, M and P indicate composite, matrix and particle, respectively. The average coefficients of thermal expansion in the temperature range from R.T. to 473 K were 19.3 $\times$ 10$^{-6}$ K$^{-1}$ for the 20 vol% SiC particle/Al-Cu composite and 15.9 $\times$ 10$^{-6}$ K$^{-1}$ for the 40 vol% SiC particle/Al-Cu composite, which were 78% and 65% of that of matrix alloy, respectively. The observed values were close to the theoretical ones based on the rule-of-mixture and Shapery’s model, suggesting that a strong bonding of the SiC particle to the matrix at the interface is obtained by LPI process.

3.2 Effect of thermal cycling on the microstructure of PRMMC

Figure 4 shows the microstructure of the 20 vol% SiC particle/Al-Cu alloy composite before thermal cycling test. The particle distribution seems to be homogenous and bonding defects at the interface between the SiC particles and the matrix are hardly observed. As shown in Fig. 4(b), a slight degradation of SiC particles was seen on the surface of SiC particle in both the 20 vol% and 40 vol% SiC particle/Al-Cu composites. The degradation of SiC particles was considered to result from a reaction between aluminum alloy melt and SiC particles during infiltration. A reaction layer of

Fig. 2 Expansion-contraction behavior of the (a) 20 vol% and (b) 40 vol% SiC particle/Al-4 mass%Cu composites in thermal cycling test.

Fig. 3 The relationship between the coefficient of thermal expansion and particle volume fraction for the SiC particle/Al-4 mass%Cu composite.
less than 1 μm thickness was observed at the interface between SiC particle and matrix in the TEM microstructure shown in Fig. 4(c). This reaction layer was identified as Al₄C₃ by TEM-EDX. It has been reported that in the case of aluminum matrix composite reinforced with SiC particles, the reaction between molten aluminum and SiC particles results in the formation of Al₄C₃ at the interface during infiltration process, where the reaction is expressed as follows:

\[ 4\text{Al} + 3\text{SiC} \rightarrow \text{Al}_4\text{C}_3 + 3\text{Si} \]  

(5)

The θ phase crystallized preferentially around SiC particles after solidification as seen in Figs. 4(a) and (b) and at higher magnification, the θ phase was present in touch with the reaction layer of Al₄C₃ formed on the surface of SiC particle as seen in Fig. 4(c). It has been reported that the θ phase crystallizes at the interfacial area between matrix and reinforcement particles in the final stage of the solidification in the squeeze casting process and that the θ phase is formed on the Al₄C₃ layer arising from a reaction between aluminum alloy melt and TiC particles. Leon et al. have reported that the θ phase have an excellent bonding characteristic to TiC particle through Al₄C₃ resulting from the reaction between the aluminum alloy and TiC particles since the interfacial energy between the θ phase and Al₄C₃ is lower than that between α-Al and Al₄C₃. Tham et al. have reported that a strong interface bonding results from the tendency for the Al₄C₃ reaction layer to form a semi-coherent interface with the aluminum matrix. Thus it is suggested that the θ phase would form a coherent interface with the reaction layer of Al₄C₃. In the present work, the interface structure observed in as prepared specimens mainly consists of SiC, Al₄C₃, θ phase and α-Al in order, and, in part, consists of SiC, Al₄C₃ and θ-Al in order. From the viewpoint of the thermodynamic stability, the former interface structure is considered to be more stable than the latter.

Figure 5 shows the microstructures of the 20 vol% and 40 vol% SiC particle/Al-Cu alloy composites after thermal cycling test. No significant change was observed in the microstructure of both specimens. Figs. 5(c) and (d) show the TEM microstructure of the SiC particle/matrix interface in the 20 vol% SiC particle/Al-Cu alloy composite after thermal cycling test. Each phase was identified by TEM-EDX and electron diffraction patterns. The top left phase corresponds to SiC particle and the bottom right phase to α-Al. No detachment was observed at the interface. The interface structure observed in the specimens after thermal cycling consists of SiC particle, the Al₄C₃ layer, θ phase and α-Al in order, which is the same as that before thermal cycling, and is considered to exhibit a strong bonding of SiC particles to matrix through the Al₄C₃ phase and θ phase due to the coherent adhesion of SiC particles to Al₄C₃ and the strong bonding of Al₄C₃ to θ phase. In addition, the rough surface of SiC particles resulting from the reaction between the alloy melt and SiC particles during infiltration (Fig. 4(b)) remains after thermal cycling as seen in Fig. 5(b). This rough surface of SiC particles was expected to contribute to an additional enhancement of the bonding strength at the interface between matrix and SiC particles due to a mechanical interlocking to both these phases. Consequently, the above-mentioned two factors might lead to the stable thermal expansion behavior shown in Fig. 2.

### 3.3 Evaluation of bonding strength of the interface by vickers microhardness test

In order to evaluate the bonding strength of reinforcement particle/matrix interface, Vickers microhardness (HV) of reinforcement particles was measured for the 20 vol% SiC particle/Al-Cu alloy composite before thermal cycling test. Figure 6 shows an example of the results of Vickers microhardness test for the specimens with a strongly bound SiC particle/matrix interface and a weakly bound interface.
Murty et al.\textsuperscript{15} have reported that the stronger the reinforcement particles bond to alloy matrix, the higher the Vickers hardness becomes. However, in the present work, the hardness values for the specimen with a strongly bound interface are almost constant and a little higher than the intrinsic hardness of SiC (HV = 2800),\textsuperscript{16} which may be caused by “spring-back” of alloy matrix on unloading. On the other hand, the specimen with a weakly bound interface shows a large scatter in the hardness values. It is considered that when the reinforcement particles bond to alloy matrix strongly, elastic deformation of the matrix around the reinforcement particles during indentation is uniform, resulting in the constant Vickers microhardness, while in the case of a weakly bound interface, elastic deformation of alloy matrix during indentation is locally and nonuniform, that is, alloy matrix deforms near the locally bound interfaces. Such deformation may be larger due to the “point-contact” between the reinforcement particles and alloy matrix, and the amount of deformation may depend on the extent of locally bound interface, which may result in a large scatter in the hardness values.

Figure 7 shows examples of thermal cycling behavior for the 20 vol\% SiC particle/Al-Cu alloy composites with a strongly bound interface and a weakly bound interface. The coefficient of thermal expansion of the specimen with a strongly bound interface is almost constant during thermal cycling, while in the specimen with a weakly bound interface, the coefficient of thermal expansion is larger than that of the specimen with a strongly bound interface and increases with increasing the number of thermal cycling. These unstable expansion behavior for the specimen with a weakly bound interface are explained as follows. When detachment at the interface was occurred during thermal
cycling, the thermal expansion of alloy matrix couldn’t be suppressed by SiC particles. With increasing the number of cycles, the portion of the detached interface increases, leading to an increase in the coefficient of thermal expansion, approaching to that of the alloy matrix.

From the correlation between the results of thermal cycling test and Vickers microhardness test, it is suggested that the bonding strength of the reinforcement particle/matrix interface could be evaluated qualitatively from Vickers microhardness test on reinforcement particles.

4. Conclusions

(1) The coefficient of thermal expansion hardly changed and no serration was observed in the expansion-contraction curves during thermal cycling test, indicating that no detachment occurred at the SiC particle/matrix interface in both 20 vol% and 40 vol%SiC particle/Al-Cu composites.

(2) The average coefficients of thermal expansion in the temperature range from R.T. to 473 K were $19.3 \times 10^{-6}$ K$^{-1}$ for the 20 vol% SiC particle/Al-Cu composite and $15.9 \times 10^{-6}$ K$^{-1}$ for the 40 vol% SiC particle/Al-Cu composite, respectively. These values were close to the theoretical ones based on the rule-of-mixture and Shapery’s model.

(3) In the microstructure of both 20 vol% and 40 vol%SiC particle/Al-Cu composites before thermal cycling test, the reaction layer of less than 1 µm thickness was observed at the SiC particle/matrix interface. The reaction product was identified as Al$_4$C$_3$ resulting from the reaction between the alloy melt and SiC particle during infiltration.

(4) The $\theta$ phase was observed on the surface of the reaction layer of Al$_4$C$_3$ before thermal cycling test. This indicates that $\theta$ phase would form a coherent interface with Al$_4$C$_3$ reaction layer formed on the SiC particles and that the interfacial structure consisting of SiC, Al$_4$C$_3$, $\theta$ phase and $\alpha$-Al in order would exhibit a strong bonding of SiC particles to matrix.

(5) The correlation between the results of Vickers microhardness test and thermal cycling test suggests that the bonding strength of reinforcement particles/matrix interface could be evaluated from Vickers microhardness test on reinforcement particles.

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