

FABRICATION CONDITIONS OF CARBON FIBER ALUMINUM COMPOSITES BY LOW PRESSURE INFILTRATION

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Abstract

The unidirectional coal-tar pitch based carbon fiber (CF) reinforced Aluminum (Al) composites were fabricated by using low pressure infiltration process. In order to degrade the infiltration pressure and reactivity between CF and Al, copper (Cu) particle was introduced in CF preform by spark sintering. Dense composites were obtained by controlling the size of Cu particle in CF preform under 0.8MPa in applied pressure of LPI process. The TC of these composites at room temperature was about 273W/mK for longitudinal direction of CF in composites which is almost equal with theoretical value.

1 Introduction

Recently, the demands for heat-sink (heat spreader) with over 400W/mK in thermal conductivity (TC) have been spread on the field of light emitting diode (LED) and power devices. Especially, the development of this high performance heat sink is required for LED lighting fixtures with high brilliance, because LED has higher brightness at lower temperature. The demands for these heat-sinks are high TC, and adequate thermal expansion, which is almost equal to that of semi conductor and electronic device. Furthermore, the control of thermal conducting direction and high mechanical properties are required for the heat-sink. One of the suitable solutions for the development of these materials is to apply the metal matrix composites (MMC). On the other hand, as the variance of shape and size, and cost performance is important for the practical use, the developments of material system and material processing are required. It seems that suitable materials for satisfying above conditions are carbon /aluminum (Al) composites. Many carbon materials such as diamond, carbon nano-fiber, pitch based fiber, graphene and graphite has high TC and low thermal expansion. By combining aluminum with these carbon materials, it seems that we will obtain the high performance heat-sink.

On the other hand, carbon has low wettability for molten Al, and the blend of the carbon and aluminum is difficult because of big difference of the density. Thereby, the composites have been usually fabricated by only sintering process and squeeze process. Unfortunately, these processes have limit of cost performance, shape and size. Accordingly, we have been developing a low infiltration casting (LPI) process for the fabrication of MMC [1, 2, 3]. In this study, Cf preform for LPI process has been developed by inserting Cu particle into preform. Especially, the optimum conditions of composite preparation were examined. Finally, the thermal conductivity of the composites was estimated.

2 Experimental Procedures

The coal tar pitch based unidirectional CFs used in this study is K13D2U CFs, which have purchased from Mitsubishi Plastics, Inc. This CFs forms 11 μ m in average diameter and have high tensile strength (3.7GPa) and low CTE (-1.2ppm/K) with excellent TC (800W/mK) along the fiber axis. Copper (Cu) powders were used for bridging materials between CFs in order to fabricate porous CFs fiber preform. Cu in preform improves the wettability, and degrades the reactivity between CFs and molten aluminum in preform. In order to obtain the uniform distribution of CFs with securing the spaces of the conduit for Al flow in the unidirectional CF preform, atomized Cu powders (Fukuda metal, foil & powder Co, LTD.) with the mixture of different an average particle size of about 2.55 μ m and 11.79 μ m was used. Cu powders mixed with a dispersant such as polyethylene glycol (PEG) were dispersed into the CF bundles by rolling process. The graphite rod was rolled to the parallel direction of unidirectional CFs with Cu powders and PEG mixture on the unidirectional CF bundles. The volume ratio of CF and Cu was 3:1. The unidirectional CF preform was fabricated by spark sintering of the unidirectional CF mixture. The tailored unidirectional CF mixture in length of 10 mm was stacked and put into the cylindrical graphite mold and sintered by spark sintering method. The volume fraction of CF, Cu and PEG was 0.3, 0.1 and 0.6, respectively. The sintering temperatures were from 1073 to 1173 K during 1800 s with the constant voltage of 4 V and electrical current of form 380 to 410 A under vacuum environment of 2.7×10^{-2} Pa. During this process, PEG was evaporated completely. The microstructure and fracture surface of unidirectional CF preform was observed by a scanning electron microscope (SEM; Hitachi S-3000H2, Japan).

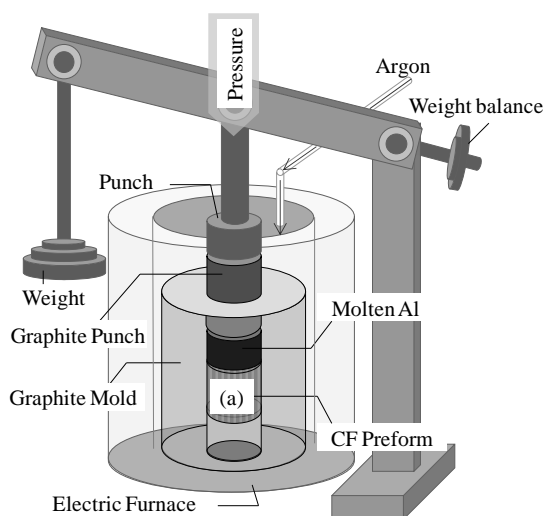


Figure 1. Schematic illustration of LPI apparatus. (a) shows the position of CF preform.

The unidirectional CF/Al composites were fabricated under the low pressure infiltration (LPI) process with 0.8MPa by means of the infiltration of molten Al (A1070) into unidirectional porous CF preform. Fig. 1 shows the schematic illustration of LPI apparatus using in this study. CF preform was set in bottom of graphite mold, and then molten aluminum was poured from upper side of mold. Pressure was applied by weight.

The microstructure and density of unidirectional CF/Al composites was estimated by a scanning electron microscope (SEM; Hitachi S-3000H2, Japan) and the water immersion method on Archimedes' principle, respectively. The thermal conductivity was evaluated by laser flash method thermal constants measuring system (TC-700, ULVAC-RICO Inc., Japan) at the room temperature in air. The specimen size of transverse and longitudinal direction of unidirectional CF/Al composites was $\phi 10 \times H 1$ and $\phi 10 \times H 2$ mm³, respectively.

3 Results and Discussion

Fig. 2 shows the microstructure of CF preform fabricated by spark sintering at 1123K. The CFs are well dispersed with maintaining the proper spaces between CFs to infiltrate the molten Al easily. Besides, there are observed two kinds Cu particles: Cu bridging particles between CFs and Cu deposition on the CF surfaces. Bridging particles affect to the strength of preform, and the compression strength of this preform was 1.67MPa, which is higher strength than applied pressure of LPI process (about under 1MPa). Cu deposition on CF forms particulate with about 1 μ m in diameter. 32.2% of CF surface area was covered with Cu deposition. In the previous studies of spark sintering process [4], the powder compact with extremely small contact area between powders proceeds mass transport by high local current densities, and occurs melt and vaporization of adjacent particles under spark sintering conditions. In case of this study, the addition of Cu powders is able to play a role of not only the Cu bridging between CFs, but also deposition on CFs by melt and vaporization under spark sintering process. It seems that Cu coatings by deposition on CFs facilitated the wettability with the molten Al.

Fig. 3 shows microstructure of CF/Al composites. CFs were distributed uniformly in composites, and casting defects such as pores were not observed. Matrix metal in composites composed of dark color and light regions are observed, which are α -Al and CuAl₂, respectively. The relative density of the composites was over 95%.

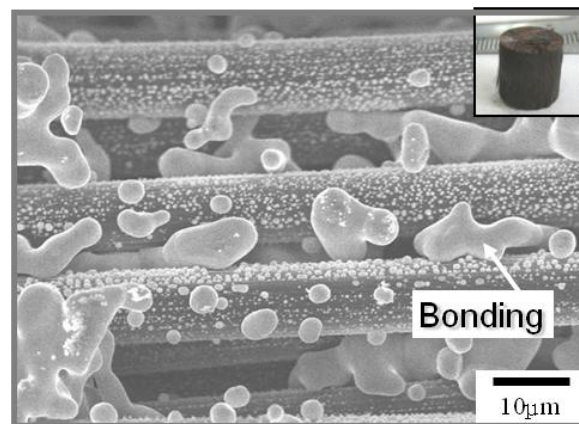


Figure 2. Microstructure of Cf preform with copper powders after spark plasma sintering.

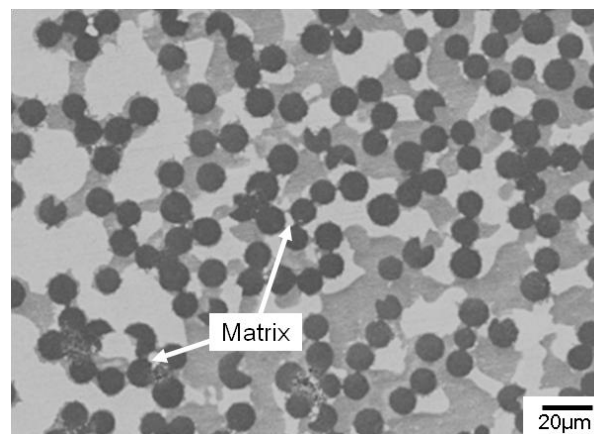


Figure 3. Cf / Al composites fabricated by low pressure infiltration method with 0.8 MPa of molten aluminum.

The TC properties of the unidirectional CF/Al composites such as matrix, longitudinal and transverse direction have been investigated, in conjunction with their theoretical values calculated by rule of mixture (ROM) of the CF and matrix. Fig. 4 exhibits the TC of the

CF/Al composites and the matrix by laser flash method at room temperature. The initial CF possessed the high longitudinal TC of about $800 \text{ Wm}^{-1}\text{K}^{-1}$ which was offered from company. Besides, the TC of the matrix such as Al- 35 wt% Cu alloy was as $71.7 \text{ Wm}^{-1}\text{K}^{-1}$ in this experiment. The 30 vol% CF/Al composites represented the excellent longitudinal TC of $273.2 \text{ Wm}^{-1}\text{K}^{-1}$. The longitudinal TC of composites was corresponding to 94 % of the theoretical value. Fig. 5 shows the schematic diagram for the heat flow into the CF/Al composites. It can be supposed that the heat was individually conducted to CFs and matrix on longitudinal direction of composites without any remarkable heat loss by fiber-matrix interface reaction. Moreover, the unidirectional CFs with high TC of $800 \text{ Wm}^{-1}\text{K}^{-1}$ is able to conduct the thermal flow most efficiently through the longitudinal direction. In the meantime, the transverse direction of CF/Al composites shows low TC of $48.0 \text{ Wm}^{-1}\text{K}^{-1}$. Although the transverse TC of K13D2U CFs was unknown from the company, that was assumed extremely low TC based on the K-1100 CFs (BP Amoco Chemicals) which had similar fiber internal structure with K13D2U CFs and represented extremely low transverse TC of $2.4 \text{ Wm}^{-1}\text{K}^{-1}$. The theoretical value of transverse TC was calculated by ROM with disregarding the transverse TC of the CFs. The experimental transverse TC was corresponding to 96 % of theoretical value. This can be explained that the heat flow was mainly conducted through the matrix. TC of this composite is almost equal to the results of ROM. In order to improve TC, the decrease of Cu content in matrix is required, and the developments of suitable fabrication conditions are required.

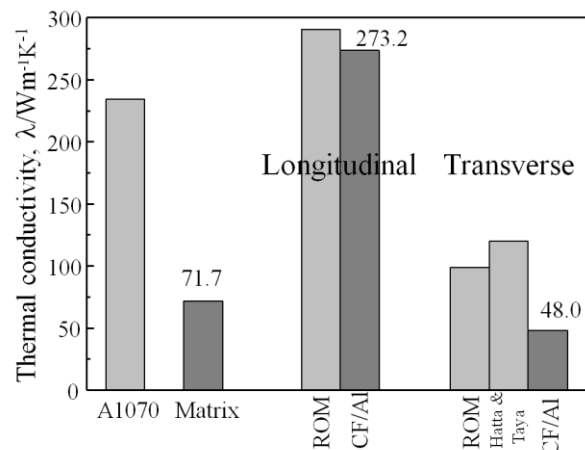


Figure 4. Comparison of experimental and theoretical results for thermal conductivity of monolithic Al and Cf/Al composites with longitudinal and transverse directions along fiber.

4 Conclusions

In order to obtain CF preform for low pressure infiltration of molten aluminum, Cu powders were inserted in preform by using spark sintering process. Cu in preform bridged CFs with high strength, and deposited on CF. Molten aluminum infiltrated into CF preform with low pressure of 0.8MPa, and 30vol% CF/Al composites with over 95% in relative density was obtained. CF was distributed homogeneously in matrix. Matrix was composed from α -Al and CuAl_2 phases. The composites represented the excellent TC of $273.2 \text{ Wm}^{-1}\text{K}^{-1}$ in longitudinal direction of fiber, which is corresponding to 94% of the theoretical value.

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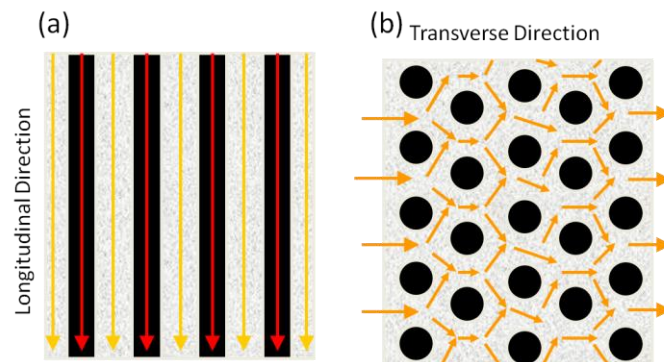


Figure 5. Schematics of heat flow in (a) longitudinal and (b) transverse directions for Cf in composites.