FABRICATION AND THERMAL CONDUCTIVITY OF CARBON NANOFIBER REINFORCED ALUMINUM MATRIX COMPOSITES BY LOW PRESSURE INFILTRATION CASTING

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ABSTRACT

Low pressure infiltration method to fabricate carbon nanofiber reinforced aluminum matrix composites with high thermal conductivity has been developed in recent years. This method requires a porous material as preform with high porosity and a network structure. In this study, vapor-grown carbon fibers (VGCFs) as reinforcement fabricated a network structure by mesophase pitch (MP) as binder in preforms. Porosity of VGCFs preform from 75.09%, 83.76% and 91.51% was controlled by spacer method. VCGFs formed a network structure by MP in preforms. Compressive property and thermal property of VGCFs preforms were investigated. The compressive yield strength of VGCF preform with 83.76% porosity and 75.09% porosity were over 0.1 MPa, which were suitable to fabricate composite by low pressure infiltration. The thermal conductivity (TC) of VGCFs preforms decreased with porosity increasing. In addition, to preform was carried on carbonization at 1500°C in Ar. By the carbonization, the compressive yield strength and TC of VGCF preform with 75.09% porosity was improved by the crystallization of MP. The preform was carried on nickel electroless coating. Then VGCF/Al composite was fabricated by nickel electroless coated VGCF preform with 75.09 % porosity at 937 K in Ar. The microstructure of fabricated composite was observed. But pores were formed in the composite for the morphology of preform surface and agglomerated MP and VGCF.

1 INTRODUCTION

Due to the increasing requirement of electronic packaging and thermal management materials, the development of metal matrix composites with high TC has been obtained in recent years, especially the aluminum matrix composites for their high thermal conductivity and low density [1-3]. The previous research utilized long carbon fibers (CFs) to manufacture CFs/Al composites with anisotropy [2]. When this composite used as heat sink which attached to substrate of high power LED, the TC of vertical substrate orientation is obviously improved. However, the thermal exchange between heat sink and air is inefficient for anisotropy. And dispersion of short carbon fibers in matrix with random direction make composites have isotropic to solve this problem [3]. For the further improvement of TC, continues fiber system and high volume fraction of CFs were also required [3]. Therefore, it's hoping that fabricate CFs/Al composites with high TC and isotropic by continues fiber system. The low pressure infiltration process has been focused to become one of the most significant method of production of CFs/Al composites for simple to fabricate with complicated shape [2, 4]. The porous material as preform is necessary in this process. And the preform requires a compressive strength over 0.1 MPa for using low pressure infiltration process and continues carbon fibers [2, 4]. However, there is no studies about microstructure, porosity, mechanical property and thermal property of preform with high porosity and composites which were fabricated by them. Due to their outstanding mechanical and thermal properties, the VGCFs [5] are considered as an ideal reinforcement with Al matrix to fabricate CFs/Al composite with high TC.

In this study, VGCFs preforms with various porosity were fabricated and investigated the

microstructures, porosity, mechanical property and thermal property. Finally, VGCF/Al composite was fabricated by low pressure casting and observed the microstructure.

2 MATERIALS AND EXPERIMENTAL PROCEDURE

VGCF with diameter of 150 nm, length of 10-20 μm (Showa Denko Co., Japan) and Mesophase pitch (MP) of diameter of 2 μm (JFE Chemical Co., Japan) were used to the fabrication of preform. NaCl particles with size of 180-360 μm were used as the spacer material to controlled pore size and porosity. VGCFs, MP and NaCl were mechanically mixed for 10 min using stirring blade. Mixture was put into a graphite mold, and pressed at 60 MPa. Molded mixture was sintered at 823 K for 5.4 ks. Finally, sintered compact was immersed into distilled water to dissolve NaCl. To investigate the effect of porosity, samples were fabricated under conditions of volume fraction of NaCl particles 70 vol.%. 80 vol.% and 90 vol.%, MP: VGCF=7:3 (vol.%). In addition, MP can be carbonized under condition of high temperature [6, 7]. To investigate the effect of carbonization, the VGCFs reform (70 vol.% NaCl and MP: VGCF=7:3) was carbonized at 1773 K for 3.6 ks in argon by a carbonization furnace.

To improve the wettability of preform, the VGCFs reform (70 vol.% NaCl) was carried on nickel electroless coating. First, the preform was defatted by acetone, then was etched by 10 vol.% HNO_3 solution for 300 s. Next step was sensitized by 4 wt% $SnCl_2 \cdot 2H_2O$ solution for 0.3 ks and activated by 0.4 wt% $PdCl_2$ solution for 300 s. Finally, the preform was set in electroless nickel bath at 338 K for 300 s. Then the VGCF/Al composites were fabricated with the coated VGCFs preform and A1070 as matrix by low pressure casting method at 1073 K in Ar under applied pressure of 0.8 MPa [4]. A scanning electron microscope, (SEM; JEOL JXA8900; 15 kV) was used for microstructure observation of the VGCFs preform. The porosity of preforms was characterized by Archimedes method [8]. In the process of infiltration, preform would be deformed and cracked. Therefore, the compressive yield strength and elasticity modulus of preform are important. The compressive properties of preforms were characterized by compression test. The compression tests were carried out using the specimens with a diameter of 10 mm and a length of 10 mm by the JIS H7902-2008 at a crosshead speed of 0.5 $mm \cdot min^{-1}$. And thermal conductivity of samples was characterized by the steady state method [9]. The VGCF preform was settled in furnace, then infiltrated with molten Al at 937K in Ar. The microstructure of fabricated VGCF/Al composite was observed.

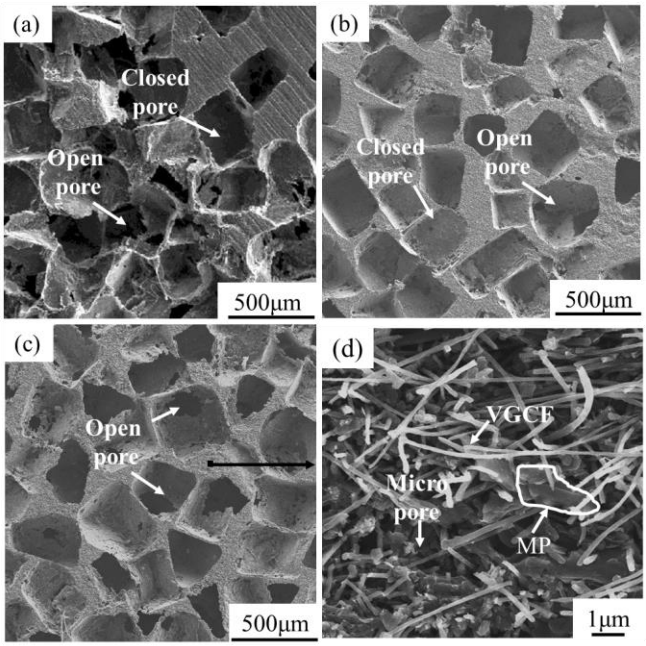

Figure 1: Macro-pores of VGCFs preform fabricated with MP : VGCF = 7 : 3 and different volume fraction of NaCl particles before carbonation: (a), (b) and (c) low magnification of 70 vol.%, 80 vol.% and 90 vol.% NaCl particles, respectively; (d) high magnification of frame of preform with 70 vol.% NaCl particles.

3 RESULTS

3.1 Microstructures of VGCFs preforms

Figure 1 shows SEM images of preforms with different porosity. As shown in (a), (b) and (c) of Figure 1, macro-pores were formed by dissolved NaCl particles. The area of pores increased with increasing volume fraction of NaCl particles. In addition, some macro-pores were isolated which formed closed pores and part of macro-pores were mutual connected which formed continues pores. It indicated that open macro-pores and closed macro-pores simultaneously appeared in the VGCFs preforms. With increasing volume fraction of NaCl particles, open pores increased and closed pores decreased. However, pores were agglomerated under condition of 70 vol.% NaCl as left part of Figure 1 (a). Figure 1 (d) shows SEM image of frame of VGCFs preform fabricated by 70 vol.% NaCl and MP : VGCF = 7 : 3. VGCFs were crossed with each other in randomly direction, and connected by sintered MP. MP powders were agglomerated and enclosed VGCFs, whereas very little MP was attached to the VGCF surface. However, micro-pores between VGCFs were observed. For the existing of micro-pores, the porosity of preforms was higher than the volume fraction of NaCl particles.

The porosity of preforms is shown in Figure 2. The porosity of VGCFs preforms, 70 vol.%, 80 vol.% and 90 vol.% of NaCl particles was 75.09 %, 83.76 % and 91.51 % respectively, and there is a linear relation between volume fraction of NaCl and porosity of preforms. The equation is as follow:

$$Porosity = 0.821 \times (Volume\ fraction\ of\ NaCl\ particles) + 17.78 \quad (1)$$

This equation indicated that the porosity of preform can be controlled by the volume fraction of NaCl particles.

3.2 Compressive strength of VGCFs preforms

Figure 3 shows compressive stress-strain curves of VGCFs preform (MP : VGCF = 7 : 3) with different porosity. The curves of preforms with 75.09 % and 83.76 % porosity exhibits the three regions: elastic region characterized by elastic modulus; stress plateau region characterized by a shallow slope corresponding to the plastic yielding and crack of pores' frame; and densification region characterized by a relatively steep slope [10]. The compressive strain was visible lower than the porosity of preform, which means part of pores remained at end of densification region. Addition, the jitter of the curve owes to the poor ductility of sintered MP, which lead to crack occurred in compressive process.

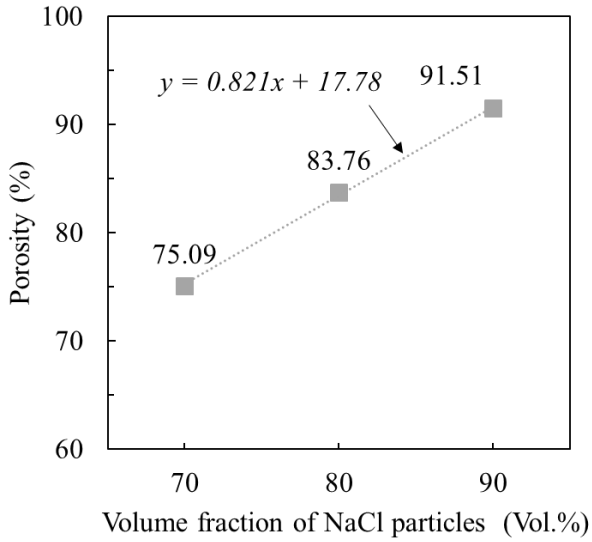

Figure 2: Porosity and volume fraction of NaCl particles (MP : VGCFs = 7 : 3).

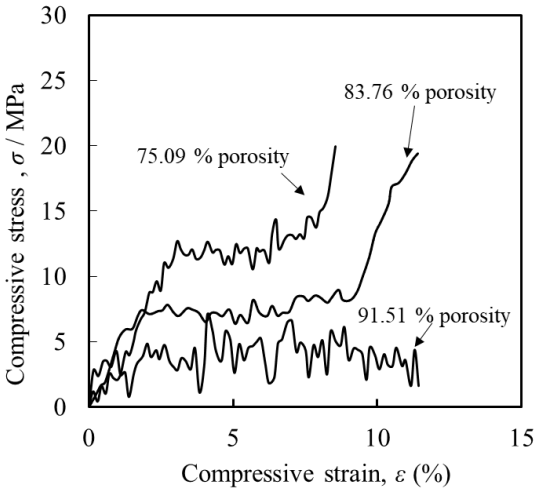

Figure 3: Compressive stress–strain curves of VGCFs preforms (MP : VGCFs = 7 : 3) with different porosity.

Normally, the compressive strength is decreasing with the increasing porosity [10, 11]. From the Figure 3, preform with 91.51 % porosity was failure for the low compressive strength as reason of high porosity. The compressive yield strength of VGCF preform with 83.76 % porosity and VGCF preform with 75.09 % porosity were 12.07 MPa and 7.41 MPa respectively. But when porosity was 75.09 %, the macro-pores were gathered together rather than uniformly distributed lead to part of poor compressive strength, and result in lower compressive yield strength.

3. 3 Thermal conductivity of VGCFs preforms

Figure 4 shows thermal conductivity of VGCFs preforms (MP : VGCFs = 7 : 3) before carbonization with different porosity and porous aluminum alloy 6101 [12]. The increase in porosity is connected the decrease in the thickness of pore's frame and increase in void fraction of whole material. The TC of air is 0.023 $W \cdot m^{-1} \cdot K^{-1}$, which means the most heat was transferred by pore's frame. Therefore, the higher porosity means less pathway for heat to transfer, than led to the low TC. Moreover, with same porosity and pore size, VGCFs preform shows higher TC then aluminum alloy 6101. Thus, in theory, VGCFs preforms can improve the TC of Al alloy matrix composite as reinforcement.

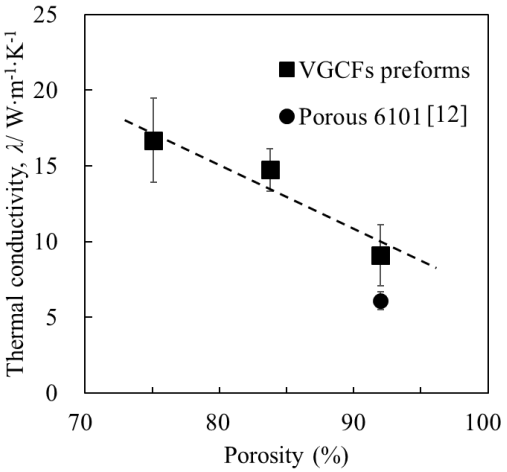

Figure 4: Thermal conductivity of VGCFs preform (MP : VGCF =7 : 3) before carbonization with different porosity and porous aluminum.

3.4 Effect of carbonization on VGCFs preforms

VGCFs were crosslinked by the bridging MP. Moreover, the VGCF surfaces were covered with a MP layer, which formed the heated MP at 823 K, and the VGCF-MP interface was consisted of linear, wave and amorphous-like structures [7]. After carbonization, the area of amorphous-like structure was decreased. It proved MP was further crystallized by carbonation. Though carbonization, the compressive yield strength of VGCFs preform was increased from 7.41 MPa to 9.32 MPa. And elastic modulus was increased from 0.7 GPa to 1.4 GPa respectively. It proved the compressive yield strength and elastic modulus of VGCFs preform were improved apparently by carbonization. After carbonation, MP was carbonized and transformed to wave-like structure. But, the TC of preform was slightly improved from 16.79 $W \cdot m^{-1} \cdot K^{-1}$ to 17.77 $W \cdot m^{-1} \cdot K^{-1}$. After carbonization, the pores remained in the preform lead to the TC was almost same with before carbonization.

3.5 Microstructures of VGCF/Al composite

Figure 5 shows the microstructure of VGCF/Al composite by using nickel electroless coated VGCF preform of 75.09 % porosity. The Figure 5 (a) shows molten Al was infiltrated inside preform by low pressure infiltration. However, pores between matrix and surface of preform were observed in the composite in Figure 5 (b). The Figure 5 (c) is high magnification image of Figure 5 (b), which shows pores at the interface of matrix and preform. The reason of this defect was fibers agglomerated into clusters, and the pressure was insufficient to infiltrate the molten Al into the gap between VGCFs clusters. Schematic diagram of Figure 5 (c) was show in Figure 6 (a). In addition, to observe the inside and surface of VGCFs cluster, Figure 5 (d) is high magnification image of Figure 5 (b). The defects of VGCF/Al composite were formed by defect in the cluster and uneven surface of cluster. Schematic diagram of Figure 5 (d) defected was show in Figure 6 (b). Therefore, to inhibit the defects of VGCF/Al composite, the dispersion of VGCFs and increase of volume fraction of MP were necessary.

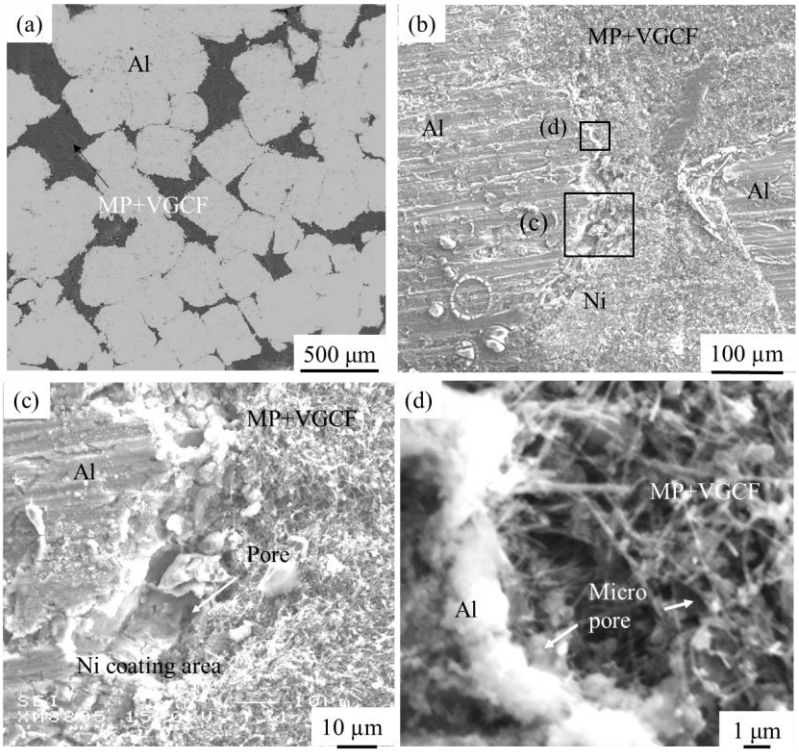

Figure 5: Microstructure of VGCF/Al composite with nickel electroless coated VGCF preform of 75.09 % porosity: (a) BSE image of longitudinal section of the VGCF/Al composite; (b), (c) and (d) high magnification SEM images of interface of VGCF/Al composite.

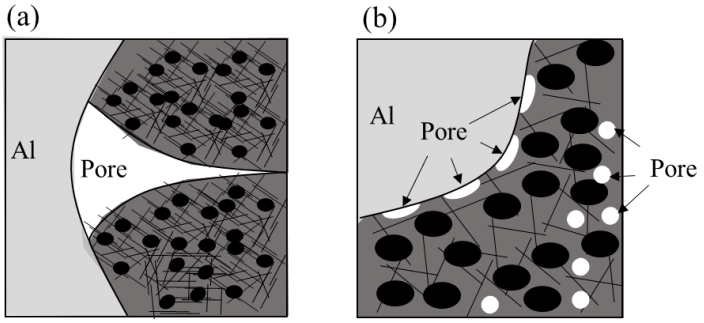

Figure 6: Schematic diagram of pores in VGCF/Al composite.

4 CONCLUSIONS

A low-pressure infiltration casting method for fabricating a VGCF/Al composite was developed. The results are summarized below.

VGCFs preform with porosity from 75.09 %, 83.76% and 91.51 % porosity was fabricated. VCGFs formed a network structure by MP as binder in preforms. VGCFs preform with 91.51% porosity was unsuited to fabricate VGCF/Al composite for the low compressive yield strength. The compressive yield strength of VGCF preform with 83.76% porosity and 75.09% porosity were over 0.1 MPa, which were suitable to fabricate VGCF/Al composite by low pressure infiltration. The TC of VGCFs preforms decreased with porosity increasing, due to the low TC of air. By the carbonization, the compressive yield strength and TC of VGCF preform with 75.09% porosity was improved by the crystallization of MP from. VGCF/Al composite was fabricated by nickel electroless coated VGCF preform with 75.09 % porosity. But pores were formed in the composite for the unevenness of preform surface and agglomerated MP and VGCF. Thus, to inhibit the defects of VGCF/Al composite, the dispersion of VGCFs and increase of volume fraction of MP were necessary.

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