

Structure Formation During Processing Short Carbon Fiber-Reinforced Aluminum Alloy Matrix Composites

S. Ciby, B.C. Pai, K.G. Satyanarayana, V.K. Vaidyan, and P.K. Rohatgi

Nickel- and copper-coated, as well as uncoated, short carbon fibers were dispersed in melts of aluminum or aluminum alloys by stirring followed by solidification of composite melts. Microstructural examination of cast composites indicated extensive damage to the surface of the carbon fibers when uncoated carbon fibers were introduced into the melt under the conditions of the present investigation. When nickel- or copper-coated carbon fibers were used to make composites under similar conditions, the fibers generally did not exhibit observable amounts of fiber surface degradation at the interface, except for small islands of an Al_4C_3 phase. When nickel-coated carbon fibers were used to make composites, the coating reacted with the melt, and $NiAl_3$ intermetallic phase particles were observed in the matrix away from the fibers, indicating a preference for nucleation of $NiAl_3$ away from the fiber surfaces. Under a transmission electron microscope (TEM), the $NiAl_3$ phase was not observed on the surface of carbon fibers, except in some regions where the $NiAl_3$ phase engulfed the carbon fibers during growth. When copper-coated carbon fibers were used to make composites, the coating reacted with the melt, and particles of $CuAl_2$ intermetallic compound were generally dispersed in the matrix away from the fibers, except for a few locations where the $CuAl_2$ phase was found at the interface under TEM observation. These microstructures are discussed in terms of nucleation of primary α aluminum and $NiAl_3$ or $CuAl_2$ phases and the interaction between short carbon fibers and these phases during growth while the composite was solidifying. Additionally, the role of the reaction between nickel or copper coatings and the melt on structure formation is discussed; some of the differences between the nickel and copper coatings are attributed to the fact that nickel dissolves with an exothermic reaction. The differences between solidification of short fiber composites and particle or fiber composite are also discussed.

Keywords

aluminum, metal matrix composites, short carbon composites, solidification

1. Introduction

THE structure and properties of cast aluminum alloy matrix composites are largely influenced by the size, shape, volume fraction, physical, chemical, and surface properties of the reinforcements, and the alloy chemistry of the matrix as well as the conditions prevailing during the solidification of the composite. The contribution of some of the above-mentioned factors on the structure and properties of the particulate and long-fiber composites have been discussed previously.^[1-3] Important phenomena that occur during the solidification of the composite are (1) nucleation and growth of phases and the role of reinforcements on both nucleation and growth, (2) effect of the movement of the solid/liquid interfaces on the redistribution of the reinforcements, (3) effect of gravity-induced settling or flotation of reinforcement on the distribution of reinforcement, (4) chemical interaction of the reinforcement with the liquid before and during solidification, (5) effect of convection currents and other hydrodynamic phenomena before and during solidification, and (6) effect of reinforcements on thermophysical and

fluid behavior properties of the matrix that influence solidification, for example, latent heat, thermal conductivity, and viscosity.

Reports are available on structure formation during the processing of continuous carbon fiber-reinforced aluminum alloy matrix composites,^[4,5] silicon carbide particulate composites,^[6,7] aluminum alloys with different reinforcements processed under varying conditions,^[8] including microgravity conditions.^[9] In most of these studies, the reinforcements are either continuous fibers or particulates. The present article concentrates on short-fiber composites that are expected to have unique properties in terms of enhanced modulus and strength and almost isotropic behavior. The solidification of short-fiber composites is assumed to be different from that of long fibers, because the solidification and hydrodynamic behavior of the melt is likely to be affected by the larger aspect ratio of the fiber and the higher frequency of the presence of fiber ends, which have a different type of surface. In earlier works, the authors have discussed some aspects of the synthesis of aluminum-base composites using a stir casting technique with either nickel-^[10] or copper-^[11] coated short carbon fibers. This article discusses the structure formation during processing of these composites.

2. Experimental

Chopped (1 to 3 mm length) as well as continuous rovings of the T-300 grade carbon fibers (supplied by M/s. Courtaulds Grafil, U.K.) were coated with nickel and copper using cementation or electroless or electroplating techniques.^[10,11] Under optimum conditions, more than 98% of the total surface area of the fibers was coated with an average coating thickness of

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about 0.4 μm of the metal. The coated carbon fibers (5 wt% of fibers exclusive of coating) were dispersed in commercially pure aluminum matrix (Al-0.2Si-0.16Fe) using a stir casting technique that involved stirring the molten metal (at 1013 ± 5 K) with a mechanical impeller while introducing the coated fibers into the vortex and then casting the slurry into ingots after mixing. In some cases, about 2 wt% magnesium was added to the melt prior to the addition of the fibers to improve the wetting between the reinforcement and the melt. Selected experiments were also conducted with uncoated chopped carbon fibers.

The composites were examined under optical, scanning, and transmission electron microscopes (TEM) to evaluate the fiber distribution and fiber matrix interaction. Selective area diffraction (SAD) patterns of the interface as well as the matrix

of the composites were also taken to identify the crystal structures of phases present in the microstructures.

3. Results and Discussion

A typical microstructure (Fig. 1) of the uncoated carbon fiber-dispersed composite suggests fiber matrix interaction, indicated by the serrations and roughness on the fiber surfaces. In composites, where magnesium was added to the melt along with fibers, small amounts of Mg_2Si were also present at the fiber matrix interface, which contained large quantities of reaction products. It is evident from the microstructure that processing conditions used in the present study were less than optimum for dispersion of uncoated carbon fibers, because the fibers exhibited signs of reaction with the matrix at the fiber surfaces. This reaction between the surface of the carbon fibers or graphite particles and the melt generally leads to formation of large crystals of the Al_4C_3 phase, resulting in degradation of the fiber as well as of the composite.^[3-6] Any processing modification including the possible use of nickel and copper coatings, which can minimize this reaction, needs to be explored.

The optical microstructures of the composites prepared with three different types of nickel coatings (Fig. 2), deposited using the electroless, cementation, or the electrolytic method, did not reveal significant differences,^[10] even though the electroless nickel coating on the fibers exhibited improved adherence, continuity, and uniformity. The microstructures (Fig. 2) depict regions of α aluminum that are free of fibers or NiAl_3 , which form due to pushing of the short fibers by the growing α dendrites. The distribution of short carbon fibers, which appear to be randomly oriented, as evidenced by circular or elliptical cross sections and segments of varying length of fibers, is quite uniform compared to the greater segregation observed in aluminum-graphite particle compositions.^[11] The main features of the optical microstructures (Fig. 2a to c) reveal both the primary and the eutectic NiAl_3 phase; there is no preferential crowding of these phases near the fibers. Apparently, much of

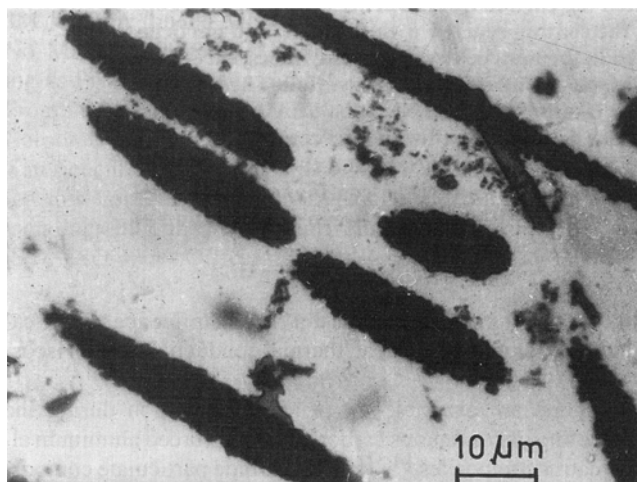


Fig. 1 Typical microstructures of aluminum matrix composite made using uncoated carbon fibers showing signs of reaction at the matrix fiber interface.

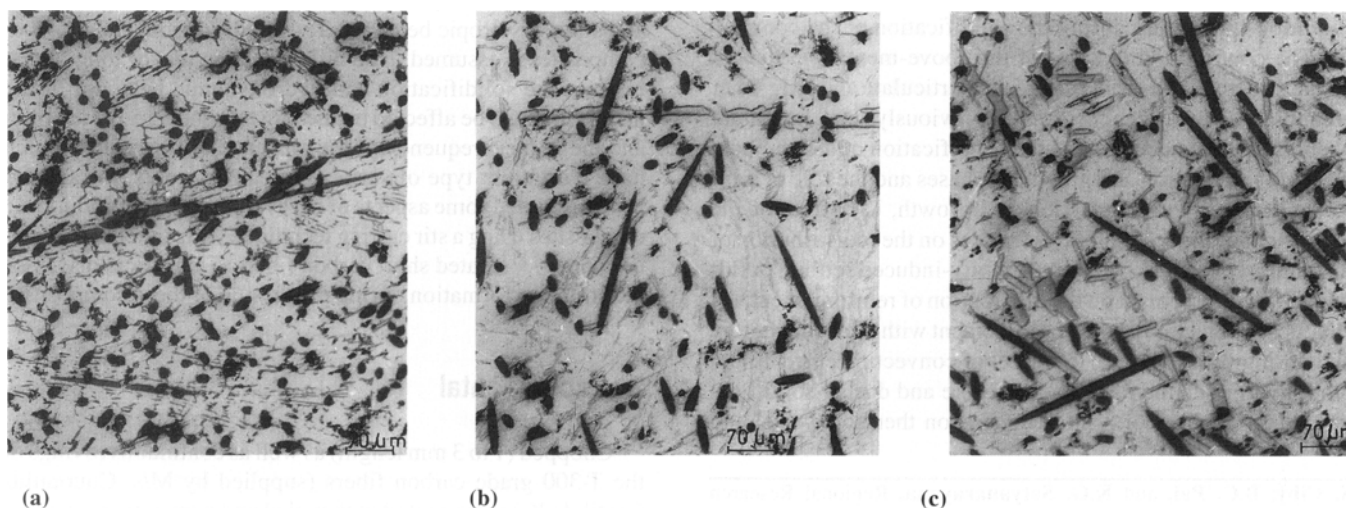


Fig. 2 Typical microstructure of composite made using nickel-coated carbon fibers. (a) Electroless coating. (b) Electrolytic coating. (c) Cementation coating.

the nickel coating dissolves in the molten aluminum alloy during stirring of the melt, and the melt containing nickel freezes, with the formation of α aluminum grains. The α aluminum dendrites reject the nickel in the interdendritic regions and simultaneously push the short carbon fibers during growth into the same regions. The enrichment of the melt by nickel in selected regions, either due to lack of diffusion of nickel dissolving from the surface of the fibers or the nickel being rejected by growing α aluminum dendrites, presumably leads to the nucleation of the primary NiAl_3 phase, which is able to grow to long, angular

crystals on the order of $50\text{ }\mu\text{m}$ in width and sometimes varying in length up to several hundred microns along with the aluminum-nickel eutectic which forms in the interdendritic regions. In several locations, the surfaces of the NiAl_3 crystals and carbon fibers are in contact, suggesting that NiAl_3 may have nucleated on the surface of carbon fibers; however, there are other locations where NiAl_3 crystals are not associated with graphite fibers (Fig. 2). In some places, carbon fibers have intercepted or are embedded in the NiAl_3 phase (Fig. 2a). This could have occurred by either nucleation and growth of NiAl_3 around graphite fibers or engulfment of sections of graphite fibers during the growth of primary NiAl_3 crystals from the melt.

In some cases (Fig. 2b), long NiAl_3 crystals appear to have grown across graphite fibers, encapsulating parts of several graphite fibers encountered during growth. This suggests that growing NiAl_3 crystals do not necessarily push away the short graphite fibers.

The dissolution of nickel or copper coatings in the melt, which is being stirred or has convection currents even after stirring has stopped, is essentially a problem of physicochemical hydrodynamics. Both diffusion and transport of atoms down the concentration gradient at the interface as well as the convective transport of the solute atoms due to the movement of liquid will determine the rate of dissolution. It is therefore necessary to consider two length scales—the hydrodynamic (Prandtl) boundary layer, thickness $\delta v = (v/uL)^{1/2}$, and the diffusion layer, thickness $\delta D = (D/v)^{1/3}$, where v is the kinematic viscosity of the liquid; u is the velocity of the liquid; D is the diffusion coefficient; and L is the linear dimension. The computations of dissolution rates show that dissolution of the coatings will occur in a very short time compared to the time scale of mixing of fibers in the liquid.

The EDAX and SAD patterns of the fiber matrix interface did not reveal the presence of a NiAl_3 phase at the interface, except in those places where a massive NiAl_3 phase is surrounding the fibers. However, in some places at the interface, small islands of Al_4C_3 were detected (Fig. 3) and confirmed by SAD patterns. This occasional presence of Al_4C_3 (Fig. 3) at the interface is in sharp contrast to the large crystallites of Al_4C_3 present all along the interface of uncoated carbon fibers and particulates dispersed in aluminum.

In composites made with copper-coated carbon fibers, using either the electroless or cementation processes^[11] for copper

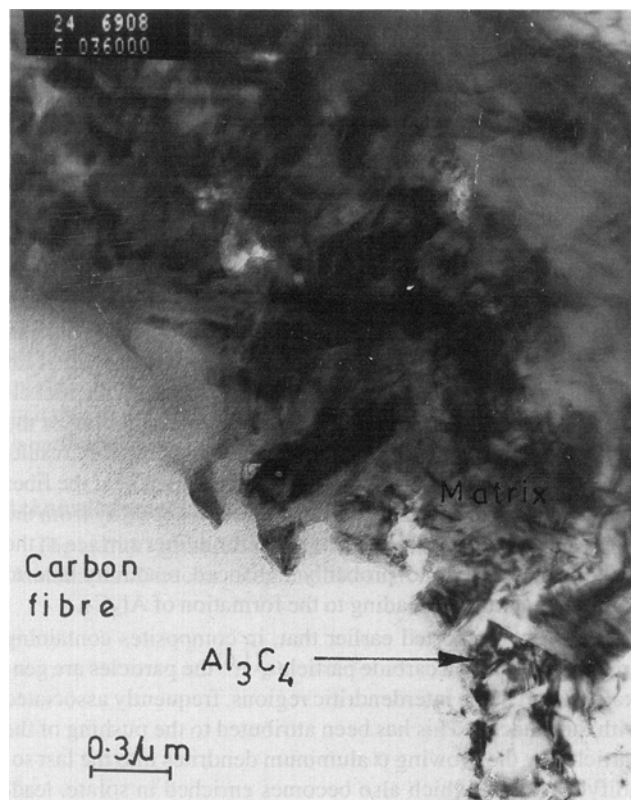


Fig. 3 Interface of carbon fiber and matrix in electroless nickel-coated fiber-dispersed composite showing Al_4C_3 formation in some places (dark-field image).

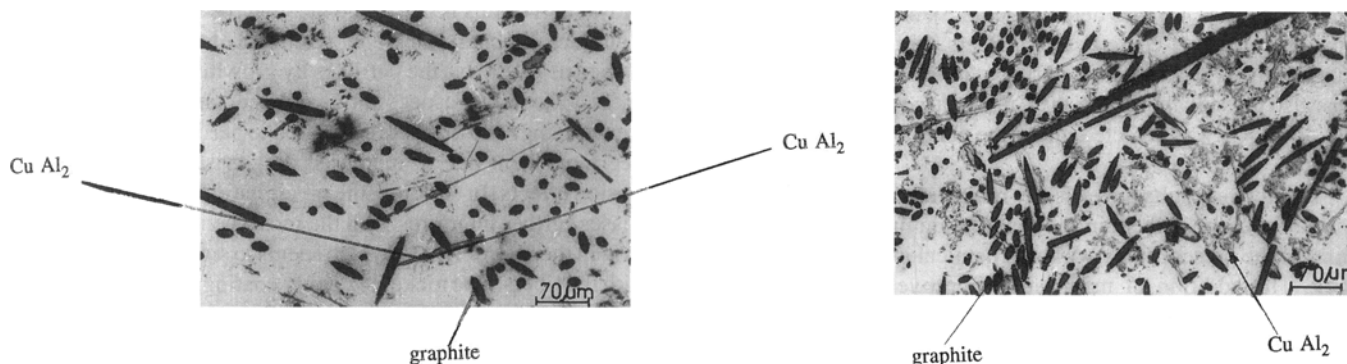


Fig. 4 Typical microstructure of composites made with copper-coated carbon fibers. (a) Electroless copper-coated fibers. (b) Cementation copper-coated fibers.

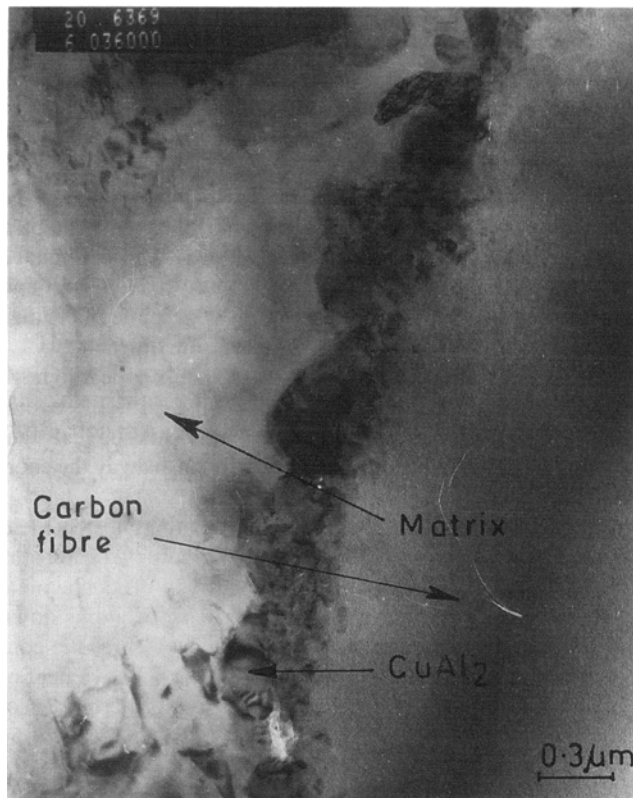


Fig. 5 Interface of carbon fiber and matrix in electroless copper-coated fiber-dispersed composite showing CuAl_2 precipitate formation at the interface.

coating, distribution of the fibers was quite similar (Fig. 4). The distribution of short carbon fibers in the microstructure is much more uniform than that observed for particulate graphite.^[1] The CuAl_2 phases were uniformly distributed in the matrix; again, no excessive crowding of these phases was observed near the fiber matrix interface. This suggests that copper was able to dissolve from the surface of the fibers, and much of it could get transported and mixed with the matrix liquid, resulting in precipitation of CuAl_2 during solidification of the composite. The SAD and EDAX analysis of the fiber matrix interface revealed the presence of the CuAl_2 phase at the interface, but no Al_4C_3 was detected (Fig. 5), as in the case of uncoated and nickel-coated carbon fibers. In composites where magnesium was also added during synthesis, the Mg_2Si phase was also present along with the CuAl_2 phase near the interface, but no Al_4C_3 was detected under the conditions of the present work.

The above results have clearly indicated that it is difficult to introduce carbon fibers in molten aluminum under the conditions of the present study without chemical interaction with the melt. The addition of magnesium improves the wetting between the fibers and the matrix and permits introduction of higher amounts of short fibers than is possible in the absence of magnesium. However, the reaction between carbon fiber and molten aluminum still occurs even when magnesium is present in the liquid. The coating of nickel over the carbon fiber ap-

pears to reduce its reaction with the molten aluminum, because no interfacial reaction is observed between the fibers and the matrix when composites are fabricated using nickel-coated fibers. The detection of Al_4C_3 at certain places at the interface with SAD analysis suggests that the reaction between the carbon fiber and molten aluminum probably occurs in isolated places of high reactivity after the dissolution of the nickel to form NiAl_3 , which is distributed in the matrix away from the fiber surface as a result of diffusion and fluid flow during mixing.

When copper-coated carbon fibers were used to make the composites under identical conditions, no Al_4C_3 was observed at the interface. Instead, CuAl_2 was detected at the interface only occasionally. Because composites using nickel- and copper-coated fibers were synthesized under similar conditions, the contact period between the melt and the fibers was the same in both cases. The occasional presence of CuAl_2 phase at the interface in copper-coated fibers can only be explained in terms of the slower rate of dissolution and lower mobility of copper in aluminum, resulting in formation of CuAl_2 at the interface. The formation of CuAl_2 as a result of the dissolution of copper in aluminum is endothermic, and the temperature near the interface of copper-coated fibers decreases during dispersion of fibers and their residence in the molten aluminum. This can decrease the mobility of copper, part of which forms CuAl_2 at the interface. In the case of composites made with nickel-coated fibers, the formation of NiAl_3 due to dissolution of the nickel coating is an exothermic process. This probably results in an increase in the local temperature of the melt near the fiber surface, leading to enhanced transport of nickel away from the fiber. This results in exposure of the carbon fiber surface to the molten aluminum and probably enhanced reactivity due to higher temperatures, leading to the formation of Al_4C_3 .

It has been reported earlier that, in composites containing graphite and silicon carbide particles,^[1,6] the particles are generally found in the interdendritic regions, frequently associated with the eutectic. This has been attributed to the pushing of the particles by the growing α aluminum dendrites into the last solidifying liquid, which also becomes enriched in solute, leading^[6] to the precipitation of eutectic in the interdendritic regions where the particles are pushed.

In the present study, the segregation of short fibers in the interdendritic regions is not as pronounced when nickel- or copper-coated fibers were dispersed in the matrix of aluminum alloys. Part of this is due to the fact that the interfiber liquid regions are probably small in size, and the fibers cannot be pushed as readily as the particles by growing α aluminum dendrites. The large aspect ratio and the much larger surface area/volume ratio of fibers compared to particulates retard their free movement due to increased drag, viscosity, and surface tension effects. In addition, the precipitation of NiAl_3 or CuAl_2 phases further interferes with the movement of fibers, particularly in the case of NiAl_3 , which seems to significantly interfere with the fibers. The nickel or copper coating, therefore, reduces the reaction between the matrix and the surfaces of short carbon fibers while improving their distribution in the matrix. In addition, the presence of NiAl_3 or CuAl_2 phases formed due to the dissolution of the coating are likely to lead to improved high-temperature strength properties of the composite.

4. Conclusions

The composites made with uncoated carbon fibers exhibited extensive reaction between the short carbon fibers and the matrix. No noticeable reaction was observed between the fibers and the matrix when nickel- and copper-coated fibers were used under similar conditions. Transmission electron microscopy of nickel-coated carbon fiber-dispersed composites revealed isolated Al_4C_3 islands at the interface. Likewise, isolated islands of CuAl_2 phase were observed at the fiber matrix interfaces of copper-coated carbon fiber-dispersed composites. Optical metallography revealed frequent interference and crossing of carbon fibers by the NiAl_3 phase; in other areas, the NiAl_3 phase either appeared by itself or on the surface of graphite fibers. Similar interaction of fibers with the CuAl_2 phases was not readily observed in copper-coated fiber-dispersed composites. The distribution of short carbon fibers in the microstructures is much more uniform compared to that of graphite particles in composites made under similar conditions. The use of nickel- or copper-coated short carbon fiber results in further improvement in the distribution of fibers compared to uncoated carbon fibers or graphite particulates. The more uniform distribution of graphite in these short fiber-reinforced composites could be due to reduced pushing of fibers during solidification and higher apparent viscosity of the composite melt due to the higher aspect ratio of the short fibers compared to particulates.

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