Fabrication of carbon fibre-reinforced aluminium composites with hybridization of a small amount of particulates or whiskers of silicon carbide by pressure casting

H. M. CHENG, A. KITAHARA, S. AKIYAMA

Government Industrial Research Institute—Kyushu, Shukumachi, Tosu, Saga 841, Japan

K. KOBAYASHI Department of Materials Science and Engineering, Nagasaki University, 1-14 Bunkyo-machi, Nagasaki 852, Japan

B. L. ZHOU Institute of Metal Research, Academia Sínica, 72 Wenhua Road, Shenyang 110015, China

An investigation was carried out on the fabrication of carbon fibre-reinforced aluminium matrix composites with hybridization of particulates or whiskers of silicon carbide by pressure casting. A small amount of particulates or whiskers was uniformly distributed among carbon fibres and the preforms prepared from the treated fibres were directly infiltrated by molten aluminium under applied stress. It was found that the longitudinal tensile strengths of hybrid composites were greatly improved, although their fibre volume fractions were very low compared to those of conventional composites. With this hybridization method, it is also practical to tailor the fibre volume fraction of composites from 60 to 25 vol %, which is not possible in direct infiltration of fibre preforms by pressure casting. The results obtained lead to the conclusion that particulate or whisker additions act not directly as reinforcements but as promoters to improve the infiltration performances of fibre preforms, and consequently to increase the strength-transfer efficiency of carbon fibres. The addition of particulates or whiskers can also improve other properties of the composites, such as hardness and wear resistance.

1. Introduction

Over the years carbon fibres (CF) have been considered as one of the most important reinforcements for aluminium and its alloys to fabricate advanced composite materials. This continuing interest in carbon fibre-reinforced aluminium (CF/AI) composites is due to their very high strength and stiffness, low density, low coefficient of thermal expansion and high thermal/electric conductivities. However, CF/AI composites also suffer from some common and critical problems, as do many other ceramic fibre/whiskerreinforced aluminium composites.

(i) Aluminium poorly wets carbon fibres [1]. As a result, the direct infiltration of liquid aluminium into the preforms of carbon fibres may be impossible or at least incomplete in pressure- or squeeze-casting processes.

(ii) Interfacial chemical reactions occur at the fibre-matrix interfaces in the manufacturing process or in elevated temperature applications, causing fibre degradation [2].

(iii) The fibre volume fraction of composites is difficult to control, especially in pressure casting, in order to be able to satisfy the various needs of different customers with the minimum fibre consumption, which means saving costly carbon fibres.

Methods of fabricating CF/Al composites have therefore generally involved the application of techniques such as metallic and ceramic coatings on the surfaces of carbon fibres and suitable alloying additions to aluminium melt, etc., in order to prohibit interactions and to enhance the wettability of carbon fibres and aluminium [3-5]. More recently, two entirely different approaches have been developed for the manufacture of CF/Al composites directly by casting. One is by pre-treatment of carbon fibres with K_2ZrF_6 [6, 7], but application of K_2ZrF_6 creates conditions which are conducive for carbide formation during liquid aluminium infiltration [8]. Another approach is based on pre-distribution of whiskers or particulates of silicon carbide among carbon fibres [9, 10]. It is reported that the longitudinal flexural strength [9] and tensile strength [10] of hybrid composites are greatly improved compared to those of conventional composites. Moreover, by using the hybridization method the fibre volume fraction of composites can be easily controlled in a wide range [10].

In the present work, unidirectional SiC particulate-

hybridized and SiC whisker-hybridized carbon fibrereinforced aluminium composites (abbreviated as SiC_p -CF/Al and SiC_w -CF/Al composites, respectively) were directly prepared by pressure casting. The effects of particulate- and whisker-addition volume fractions on the longitudinal tensile strengths, fibre volume fractions and fracture morphologies of composites obtained were investigated in detail. The functions of particulates or whiskers in the infiltration process and their contribution to the strength of achieved composites are also discussed.

2. Experimental procedure

The carbon fibre used in this investigation was of the PAN-based high-modulus type. Some properties, along with those of SiC particulates and whiskers and matrix metal of aluminium–12 wt % silicon alloy, are listed in Table I. All of the data are quoted from the manufacturers' technical specifications, except the tensile strengths of the carbon fibres and aluminium alloy which were measured directly. Tensile strength tests of carbon fibres were made on single fibres with a miniature testing machine, the number of testing was 50 with a gauge length of 25 mm at crosshead speed of $8.3 \,\mu m \, s^{-1}$. The results were evaluated by the Weibull distribution theory.

The process adopted to prepare hybrid composites included the following sequential steps.

(i) Distribution of particulates or whiskers among carbon fibres. Carbon fibres were impregnated into an aqueous suspension of particulates or whiskers, using a polymer as binding agent and an organic-metallic compound as dispersing agent. After this treatment, particulates and whiskers were uniformly distributed among carbon fibres with the binding of the binder, as shown in Fig. 1a, b, respectively, but when the whisker concentration in the suspension was high, whiskers were agglomerated in some areas because of their long length.

(ii) Carbon fibre preform preparation. The impregnated fibres were dried to a certain extent, then cut to a length of 80 mm and pre-pregged in one direction into a Shirasu Balloon (expanded volcanic glass) pre-

TABLE I Properties of the raw materials utilized

Carbon fibre	High modulus:	Diameter: 7 µm Tensile strength: 2358 MPa ^a Young's modulus: 359 GPa
SiC particulates	α type	Average diameter: 0.6 µm
SiC whiskers	β type	Diameter: 0.1–1.0 μm Length: 10–200 μm
Aluminium alloy	Al-12% Si	Tensile strength: 176 MPa ^a Melting point: ~ 853 K

* Values measured in the present investigation.

form mould to obtain a fibre preform 80 mm long, 9 mm wide and 3-5 mm thick.

(iii) Fabrication of composites by pressure casting. The preforms described above were preheated to the temperature of 650 K along with the cast iron mould, then pressure casting was carried out at a pressure of 49 MPa, a melt temperature of 1053 K and a pressurekeeping time of 60 s. Both steps were practised in the air.

All of the composites obtained were examined in the as-fabricated state. Longitudinal tensile strength was tested at room temperature by an Instron testing machine. Tensile test specimens were prepared by machining and grinding the cast billets to the geometry illustrated in Fig. 2. The surfaces of specimens were polished with emery paper of 180-1200 grit size, and tensile tests were performed at a crosshead speed of $8.3 \,\mu\text{m s}^{-1}$. The tensile fracture morphologies were also observed with a scanning electron microscope (SEM).

Fibre volume fractions of all composites prepared were measured using the point-count method. The size of a unit square of point-count mesh was 4×4 mm and its effective counting points were 360 for an optical micrograph with a magnification of $\sim \times 1000$. For every specimen, 15–20 pieces of optical micrograph were counted with the count mesh and the average fibre volume fraction was calculated. The additive volume fractions of hybrid composites were computed according to the relative volumes of addi-



Figure 1 SEM micrographs of (a) SiC particulate-distributed carbon fibres (volume of particulate to volume of fibre: 7.3%); (b) SiC whiskerdistributed carbon fibres (volume of whisker to volume of fibre: 3.0%).



Figure 2 Geometry of tensile test specimens (mm).

tives stuck on the carbon fibres in the pre-treatment and the fibre-volume fractions of composites obtained.

3. Results

3.1. Microstructures of composite billets

Fig. 3 shows the transverse cross sections of three types of composite obtained. It is apparent that the structures are quite different between hybrid and conventional composites. On the one hand, it is found from Fig. 3a, b that the fibres in hybrid composites are completely infiltrated by the matrix to yield sound billets, and fibres are uniformly scattered over the whole area with nearly no fibre contact. No voids exist between fibres and matrix aluminium. On the other hand, in the conventional composites shown in





Fig. 3c, carbon fibres are packed closely one by one. Not only do most of them make contact with the neighbouring fibres, in some areas almost perfect hexogonal packing being observed, but also at the sites close to the contacts impregnation of aluminium is incomplete, and voids exist. It is also noted that there is a much higher concentration of fibres in the conventional composites than in the hybrids. Comparing Fig. 3a and b there seems to be no big difference in microstructure except for the fibre volume fractions between SiC_p-CF/Al and SiC_w-CF/Al composites.

The SEM micrographs showing the transverse cross sections of etched composites are given in Fig. 4. The specimens were mechanically polished and chemically etched in a 5% NaOH aqueous solution for SEM observations. Matrices around the fibres and additives were preferentially soluted. There was no evidence of interfacial layers formed between fibres and matrix which could be seen by SEM. Particulates and whiskers were harmoniously distributed around the circumference of fibres, widening the spacing distances of the fibres. And it can be seen from Fig. 4 that, although the particulate volume fraction is a little higher than that of whiskers, the distribution of fibres in the SiC_p-CF/Al composite is denser than that of the SiC_w-CF/Al composite. This indicates that the whiskers can make larger spaces than the particulates.

3.2. Longitudinal tensile strength and fibre volume fraction of hybrid composites

The longitudinal tensile strengths and fibre volume fractions of the three composites are shown in Fig. 5 as functions of the volume fractions of additives distributed in the composites. In Fig. 5b, it is indicated that as additive volume fraction increases, the fibre-volume fraction of composites decreases monotonously from about 59 vol % with no additive to about 25 vol % with the additive volume fraction of more than 4 vol %, as particulates and whiskers have widened the spaces among fibres. But as illustrated in Fig. 5a, the tensile strength of hybrid composites does not have the same tendency as the fibre-volume fraction. It

Figure 3 Optical micrographs of transverse cross sections of (a) SiC_p-CF/Al composite (particulate volume fraction: 1.0 vol %); (b) SiC_w-CF/Al composite (whisker volume fraction: 1.1 vol %); (c) CF/Al composite.





Figure 4 SEM micrographs of transverse cross sections of etched (a) SiC_p -CF/Al composite (particulate volume fraction: 2.7 vol %); (b) SiC_w -CF/Al composite (whisker volume fraction: 2.2 vol %).

increases sharply at first and reaches a maximum volume of 807 MPa for SiC_p -CF/Al composite and 746 MPa for SiC_w -CF/Al composite at the additive volume fraction of around 1 vol %, which are 37 and 27% higher, even though the fibre volume fractions



Figure 5 Effects of additive volume fraction on the longitudinal tensile strength and fibre volume fraction of hybrid composites. (a) longitudinal tensile strength; (b) Fibre volume fraction. \blacktriangle , SiC_p-CF/Al; \blacksquare , SiC_w-CF/Al; \blacksquare , CF/Al.

are 32 and 39% lower, respectively, for both types of hybrids than those of conventional composite. When the additive volume fraction exceeds 1 vol %, the tensile strength goes down mainly because of the obvious decreases of fibre volume fraction. Although the same amount of particulates or whiskers is distributed in the composites, the fibre volume fractions of SiC_p-CF/Al composite are a little higher than those of SiC_w-CF/Al composite, and consequently the tensile strength of the former is also greater than that of the latter.

3.3. Fracture morphology of composites

Specimens for fractographic studies were prepared by cutting the tensile test specimens and ultrasonically cleaning the fracture surfaces in ethanol. Fig. 6 gives the SEM fractographs of the composites. In the conventional composites shown by Fig. 6a, most of the fibres can be seen to have been totally pulled out from the matrix or stripped off from the interfaces caused by interlaminar failure. However, in the hybrid cases, although many fibres have been pulled out from matrix as well, the pull-out length is much shorter than that in the conventional case, and interlaminar failure rarely exists as shown in Fig. 6b-d. And when the amount of additives increases the pull-out length decreases, as observed in Fig. 6b, c. Comparing Fig. 6c and d, it appears that the fracture behaviours are almost the same for SiC_p-CF/Al and SiC_w-CF/Al composites.

4. Discussion

4.1. Tailoring fibre volume fraction by hybridization

In the direct-casting process (unlike other processes in which one can design the fibre volume fraction of composites by arranging the volume of precursor wires in advance), colsely-packed fibre preforms prepared from multifilament bundles are further densified randomly under the applied stress during pressure infiltration, and usually samples obtained by pressure casting and squeeze casting have as high as about



Figure 6 SEM fractographs of (a) CF/Al composite; (b), (c) SiC_w-CF/Al composite (whisker volume fractions: 1.1 and 2.2 vol %, respectively); (d) SiC_w-CF/Al composite (particulate volume fraction: 2.7 vol %).

60 vol % fibre volume fraction [10, 11]. Therefore it is almost impossible to manufacture composites with a certain fibre volume fraction as required for practical applications. Nevertheless, when considering the variations of applications and reduction of cost, it is essential to satisfy property requests with the minimum fibre consumption.

From Fig. 5b, it can be seen that the fibre volume fraction of hybrid composites has a definite relation to the volume fraction of additives, and the variation is in the range of 59 to 25 vol % with the same level of tensile strength of CF/Al composite. This indicates that fibre volume fractions can be easily controlled to such an extent as to meet the requirements of various applications with the minimum consumption of costly carbon fibres.

In comparison with the conventional composite, the decreases in fibre volume fraction of hybrids do not imply a sacrifice of the properties of materials, especially when considering the case of longitudinal tensile strength. The relationship of fibre volume fractions to the longitudinal tensile strength of hybrid composites is illustrated in Fig. 7. Even for composites with about 30 vol % fibre volume fraction, which is only the half of the value of conventional composites, the longitudinal tensile strengths still keep higher values than those of conventional composites. Thus the addition

of particulates or whiskers has certainly promoted the strengthening efficiency of carbon fibres.

4.2. Functions of additives in the infiltration process

Carbon fibres without pretreatment cannot be completely impregnated with molten aluminium through pressure casting, and most fibres make contact with the neighbouring fibres as shown in Fig. 3c. One reason for this is the poor wettability of carbon fibres and aluminium; another is that carbon fibres themselves are subjected to the applied stress during pressing, causing densification of fibres and forming close-packing microstructures in which carbon fibres make contact with one another.

In the case where particulates or whiskers are uniformly distributed among fibres, despite the fact that hybridization cannot lower the wet angle of carbon fibres and aluminium, it is possible to enlarge the spacing distances between fibres to produce many capillaries in the fibre preforms, as shown in Fig. 8. When pressure casting is applied, with their high compressive strength the evenly distributed additives are capable of enduring the applied pressure and maintaining the formed fibre separation to a certain degree, to allow the liquid metal matrix and the



Figure 7 Longitudinal tensile strength as a function of fibre volume fraction for hybrid composites. \blacktriangle , SiC_p-CF/Al; \blacksquare , SiC_w-CF/Al; \blacklozenge , CF/Al.

following plastic solidifying matrix to fill these capillaries up. The greater the amount of particulates or whiskers added, the bigger the capillaries formed; as a result, the lower the fibre volume fraction of hybrid composites. Because whiskers have longer length and lower packing density, their capacity for separating fibres is much greater than those of particulates (Fig. 8a, b). This is consistent with the experimental results of the variation tendency of fibre volume fractions given in Fig. 5b.

4.3. Contribution of additives to the properties of composites

In the conventional composites, because incomplete impregnation occurs and direct fibre contacts and voids form, the interfacial bonding strength of fibre and matrix is too weak to efficiently transfer the applied stress to carbon fibres, the principal loadcarrying constituent. And as not enough metal matrix exists around the circumferences of fibres, concentrated stress caused by the weak fibre failures cannot be released by the plastic deformation of the matrix. So the stress concentration grows very quickly even at low stress levels, and causes interlaminar failures in which fibres are broken at the weakest points and then stripped off from the very weak interfaces. These phenomena, clearly observed in Fig. 6a, result in a lower strength of composites and a lower strength transfer efficiency of carbon fibres.

Hybrid composites have much higher strength than conventional composites (Fig. 5a). However, it cannot be considered that this promotion comes from the direct contribution of additives as strength reinforcements, because of their very low volume fractions. Also, there appears to be no obvious difference in strength contribution between SiC_p-CF/Al and SiC_w-CF/Al composites, although it is well known that SiC whiskers have an astonishingly high tensile strength. As mentioned above, additive distribution prohibits fibre contacts and enhances the complete infiltration of aluminium effectively, which leads to production of good billets. A certain number and length of fibre pullouts on the fracture surfaces in hybrid composites is evidence that carbon fibres have played an important role in the hybrid composites. So it can be assumed that the strength increase of hybrid composites originates from the great improvement of strengthening efficiency of the carbon fibres themselves. The strength transfer efficiency of carbon fibres in hybrid composites, which stands for the ratio of the experimental strength to the theoretical strength calculated from the rule of mixtures, is illustrated in Fig. 9 as a function of additive volume fraction. In conventional composites, fibre strength transfer efficiency is only 40%, while it reaches the maximum value of about 80% and does not fall below 70% in both types of hybrid composite. The decrease of fibre strength transfer efficiency of the composites with higher additive volume fraction is due to the existence of big agglomerated grains of additives as structural defects [10].



Figure 8 SEM micrographs of transverse cross sections of (a) SiC particulate-distributed carbon fibre preform (volume of particulate to volume of fibre: 7.3%); (b) SiC whisker-distributed carbon fibre preform (volume of whisker to volume of fibre: 7.7%).



Figure 9 Effect of additive volume fraction on strength transfer efficiency of carbon fibres of hybrid composites. \blacktriangle , SiC_p-CF/Al; \blacksquare , SiC_w-CF/Al; \blacklozenge , CF/Al.

TABLE II Vickers hardness of SiC_w-CF/Al composites

Whisker volume fraction (vol %)	1.1	2.2	2.8	
Vicker's hardness (49 N, 30 s)	58	65	69	
				_

* Measured in the direction perpendicular to fibre axis.

It is widely known that particulate- or whiskerreinforced metal composites have excellent wear resistance and other properties [13], thus it is possible to improve such properties of carbon fibre-reinforced aluminium composites with the hybridization of particulates or whiskers. For instance, it has been reported that the transverse tensile strength of hybrid composites is higher than that of conventional composites [9]. Table II gives the Vickers hardness of some SiC_w-CF/Al composites, and shows an increase of hardness as the amount of whiskers increases. Considering the positive relation of hardness and wear resistance, it indirectly suggests some improvement of wear resistance of the composites.

From the above discussion of the functions of additives, it can be seen that this hybridization method not only can be used in CF/Al composites in order to improve the strengthening efficiency of carbon fibres and to tailor the fibre volume fractions of composites, but also can be applied to other multifilament fibres such as silicon carbide fibre- and alumina fibrereinforced metal matrix, and even plastic composites, to achieve those targets.

5. Conclusions

1. Multifilament-type carbon fibre-reinforced aluminium composite materials can be fabricated directly by pressure casting with hybridization of a small amount of particulates or whiskers of silicon carbide.

2. The longitudinal tensile strengths of hybrid composites are improved greatly, although their fibre volume fractions are very low compared to those of conventional composites.

3. The addition of particulates or whiskers can also easily control the fibre volume fractions of hybrid composites, so it is practical to tailor fibre volume fractions according to application needs and therefore to save costly carbon fibres.

4. The functions of additives are mainly to improve the infiltration performance of fibre preforms, and consequently to increase the strength transfer efficiency of carbon fibres. Therefore, this method can be applied to other multifilament fibre composite systems as well.

5. The other properties, such as hardness and wear resistance, of CF/Al composites can also be improved by hybridization with particulates or whiskers.

Acknowledgements

This investigation was carried out in the Government Industrial Research Institute—Kyushu, AIST, MITI, Japan. One of us, H.M.C., is on research leave supported by a scholarship from World Laboratory— International Centre for Scientific Culture, Switzerland. We also record our appreciation of the collaboration of our colleagues, especially Dr Saitoh, Mr Ueno, Dr Hirai, Dr Sakamoto and Mr Baba, for their kind help and constructive discussions, and Mrs Hashimoto for her aid whenever necessary.

References

- 1. I. H. KAHN, Met. Trans. A 7A (1976) 1281.
- M. KH. SHORSHOROV, in Proceedings of the 4th International Conference on Composite Materials, Tokyo, 1982, edited by T. Hayashi, p. 1273.
- 3. A. A. BAKER, A. MARTIN and R. J. BACHE, Composites 2 (1971) 154.
- 4. M. F. AMATEAU, J. Compos. Mater. 10 (1976) 279.
- A. KIMURA, T. TERAOKA and R. SAGARA, in Proceedings of the 4th International Conference on Composite Materials, Tokyo, 1982, edited by T. Hayashi, p. 1451.
- 6. J. P. ROCHER, J. M. QUENISSET and R. NASLAIN, J. Mater. Sci. Lett. 4 (1985) 1527.
- 7. Idem., J. Mater. Sci. 24 (1989) 2697.
- S. N. PATANKAR, V. GOPINATHAN and P. RAMA-KRISHNAN, Scripta Metall. Mater. 24 (1990) 2197.
- S. TOWATA, H. IKUNO and S. YAMADA, Trans. Jpn Inst. Metals 29 (1988) 314.
- 10. H. M. CHENG, A. KITAHARA, K. KOBAYASHI and B. L. ZHOU, J. Mater. Sci. Lett. 10 (1991) 795.
- 11. S. TOWATA, S. YAMADA and T. OHWAKI, Trans. Jpn Inst. Metals 26 (1985) 563.

Received 4 March and accepted 1 July 1991