Advanced Composite Materials
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/tacm20

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Version of record first published: 02 Apr 2012.

To cite this article: J. Pan , S. P. Lee , M. Yoshida , G. Sasaki , N. Fuyama , T. Fujii & H. Fukunaga (2001): Effect of whisker surface treatment on the strength of Al18B4O33/Al alloy composites , Advanced Composite Materials, 10:4, 299-307

To link to this article: http://dx.doi.org/10.1163/156855101753415328

Adv. Composite Mater., Vol. 10, No. 4, pp. 299–307 (2001)

Effect of whisker surface treatment on the strength of $Al_{18}B_4O_{33}$/Al alloy composites

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Abstract—In order to increase the whisker preform strength and decrease the interfacial reactivity between $Al_{18}B_4O_{33}$ whisker and aluminum alloy, three kinds of whisker surface treatments have been carried out. They were SiO_2 binder addition, magnesium deposition coating and spinel ($MgAl_2O_4$) coating. By a low vacuum evaporation technique (under vacuum 10 Pa at 650°C for 1 h), magnesium was deposited on the whiskers uniformly. Magnesium deposition reacted with the whisker at 800°C for 0.5 h in air to create a $MgAl_2O_4$ layer (8–10 nm in thickness) on the whisker. Three kinds of surface-treated whiskers and the untreated whisker were used to reinforce AC4CH aluminum alloy by squeeze casting method. Those surface treatments could increase the strength of whisker preform and hence decreased the preform deformation during squeeze casting. However, SiO_2 binder did not prevent the interfacial reaction. Being similar to untreated whisker composite, SiO_2 added whisker composite had a deterioration of strength after T6 treatment. Magnesium coating just avoided the interfacial reaction of $Al_{18}B_4O_{33}$ whisker with the matrix but did not prevent that with the magnesium coating itself. After T6 treatment, the strength decrease due to whisker damage and the strength increase from the matrix age strengthening did not change the total strength of composites. $MgAl_2O_4$ coating existed as an effective barrier to prevent the interfacial reaction. During T6 treating, no further whisker damage occurred and the matrix was strengthened. This produced considerable enhancement of the composite strength.

Keywords: Aluminum borate whisker; Mg-deposition; spinel coating; aluminum alloy; squeeze casting; tensile strength; interfacial reaction.

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1. INTRODUCTION

Aluminum borate $Al_{18}B_4O_{33}$ (AlBO) whisker can compete with SiC whisker as a reinforcement of metal matrix composites, because it has coinparable properties and low cost [1, 2]. Aluminum matrix composites reinforced by this whisker have been used practically in automobile engines [3, 4]. But the interfacial reaction that leads to the degradation of whisker strength must be controlled for it to have wide application. AlBO whisker reacts with the magnesium included in aluminum alloy from about 520°C, and with aluminum directly at about 730°C [5, 6]. Such temperatures are required at the fabrication and heat treatment processes of composites. As a result of the interfacial reaction, heterogenous products like spinel ($MgAl_2O_4$) and γ-Al_2O_3 are formed at the whisker surface [6–9]. This leads to deterioration in reinforcement efficiency owing to the whisker consumption and whisker length shortening [10–12]. In addition, because magnesium in the matrix alloy is also exhausted by the reaction with whisker, the age strengthening of the matrix after T6 treatment cannot appear [12, 13]. Therefore, a suitable barrier to avoid the direct contact between AlBO whisker and aluminum matrix is necessary. Squeeze casting is a simple process to fabricate AlBO/Al alloy composites. Generally, an inorganic binder such as SiO_2 is added into the whisker in order to decrease the whisker preform deformation. But SiO_2 binder seems not to prevent the whisker reaction with the matrix alloy. Under these considerations, a new surface treatment method for AlBO whisker has been developed. That is, a uniform magnesium coating is deposited on AlBO whisker by vacuum evaporation of magnesium, and then a homogenous spinel ($MgAl_2O_4$) layer is formed by the reaction between this magnesium deposition and the whisker during heat treatment. This surface coating is expected not only to act as binder of the whisker preform, but also as an interfacial reaction barrier. The aim of the present work is to investigate the effect of the different surface treatments on the preform strength and the composite strength after T6 treatment.

2. EXPERIMENTAL

AlBO whisker, manufactured by Shikoku Chemical Industry (Japan), was used as the reinforcement. Pure magnesium (purity: 99.95%) and AC4CH aluminum alloy (Al-7.3Si-0.37Mg) were employed as the coating metal of whisker and the matrix alloy of composites, respectively. Magnesium coating on whisker was carried out in a vacuum furnace as shown in Fig. 1, where a piece of pure magnesium was vapourized and then deposited on the whiskers. The furnace was heated under vacuum (10 Pa). When the temperature reached 650°C, the vacuum valve was closed and the furnace chamber was in a sealed state. After heating at this temperature for 1 h, electricity supply was switched off, and the AlBO whisker cooled down together with the furnace naturally.

Mg-deposited whiskers were taken out from the apparatus and then heated in air for 0.5 h to form a spinel layer on the whisker by the reaction of the magnesium

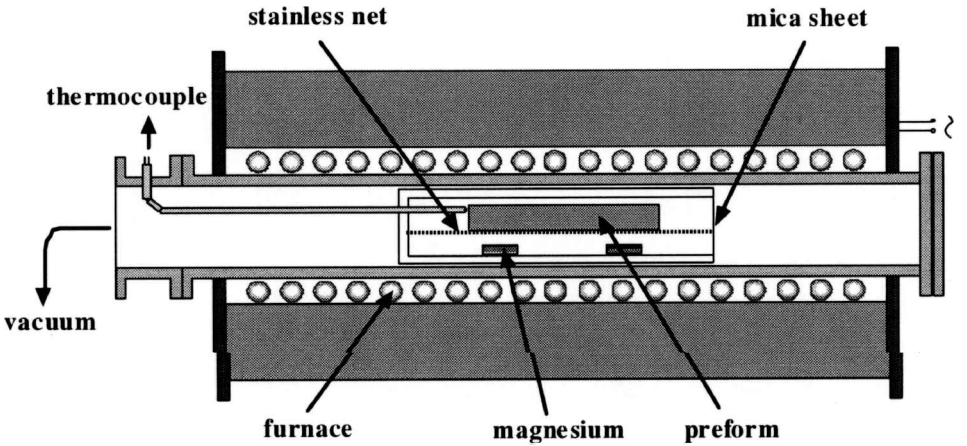

Figure 1. Schematic diagram of AlBO whisker coating apparatus.

deposition with AlBO whisker. Untreated whisker, 3 mass% SiO_2 added whisker, Mg-deposited whisker and spinel coated whisker reinforced AC4CH alloy composites were fabricated by squeeze casting, respectively. The preform preheating temperature, aluminum alloy melt pouring temperature, die preheating temperature and squeeze pressure were 700°C, 760°C, 200°C and 100 MPa, respectively. The as-cast composites were T6 treated under the following conditions: solution treating at 525°C for 8 h, water quenching and aging at 160°C for 6 h. SEM (HITACHI/ S-800), EPMA (JEOL/ JXA-9800RL) and XRD (MAC SCIENCE/ $MXP^{18}VA$) were used to identify the composition and structure of the deposition and the coatings on AlBO whiskers. Mg-deposited AlBO whiskers after heat treatment were observed using TEM (TOPCON/ EM-002B). The compressive strength of the whisker preforms and tensile strength of AlBO/AC4CH composites were evaluated with a mechanical tester (SHIMADZU/ Autograph AG-100kNG).

3. RESULTS AND DISCUSSION

EPMA analysis result of Mg-deposited AlBO whisker showed that the atomic ratio Mg : Al was 13.5 : 86.5. Since the magnesium coat was a thin layer, aluminum was detected from the whisker. From this result it could be confirmed at least that magnesium was coated on the whisker successfully. XRD patterns of the different whiskers are exhibited in Fig. 2. For the Mg-deposited whiskers, magnesium existence was hardly found (Fig. 2b). This might be attributed to the state of magnesium; it was a cluster and did not become a perfect crystal. When the Mg-deposited whisker was heated at 600°C, the peaks of magnesium and spinel were not still identifiable (Fig. 2c). But in the cases of heat treatment at 700°C and 800 °C, obvious $MgAl_2O_4$ peaks could be confirmed (Fig. 2d and 2e). For the whisker after heat-treatment at 800°C, the atomic ratio Mg : Al was 12.6 : 87.4, which was

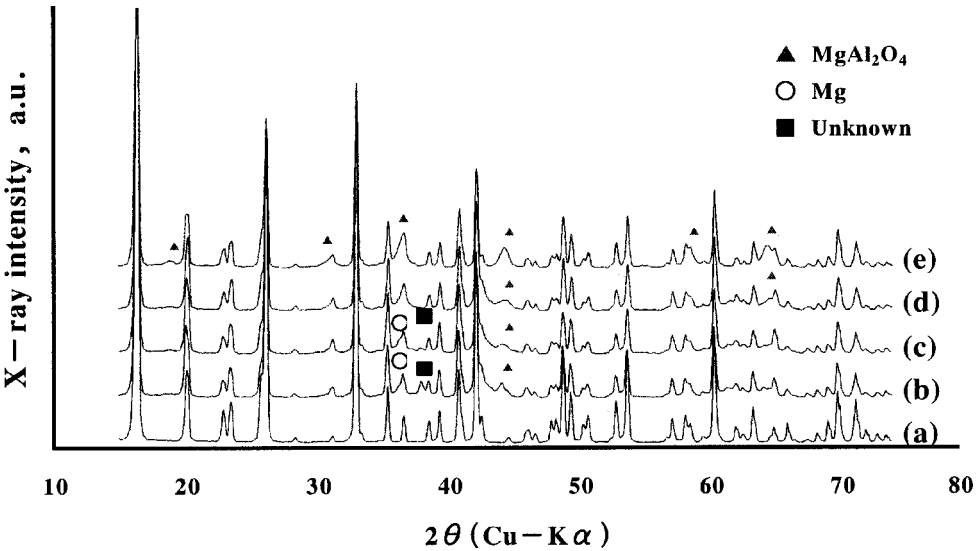

Figure 2. XRD patterns of the as received whiskers, Mg-deposited whiskers, and the Mg-deposited whiskers after heated at different temperatures. (a) As received, (b) Mg-deposited at 650°C, (c) heated at 600°C, (d) heated at 700°C, (e) heated at 800°C.

almost the same as that before heat-treatment. It is speculated that the reaction between magnesium and AlBO whisker occurs as follow:

$$4Al_{18}B_4O_{33} + 33Mg \rightarrow 33MgAl_2O_4 + 16B + 6Al \quad (1)$$

The calculation of Gibbs free energy ($\Delta G = -5.97 \times 10^6$ J/mol) indicated that this reaction can arise thermodynamically.

AlBO whiskers that coated with magnesium at 650°C for 1 h in vacuum and then heat-treated at 800°C for 0.5 h in air were observed with transmission electron microscope. Figure 3 shows a surface profile image of the whisker. A continuous reaction layer with a thickness of 8–10 nm is covering the whole whisker. It adheres to the whisker maintaining a specific crystal orientation. The identification shows that it is a spinel phase $MgAl_2O_4$. This result agrees with those from EPMA and XRD. Such a spinel layer bonded with AlBO closely may become a good barrier to prevent the reaction of the whisker with the matrix alloy. In the outside of the spinel layer, MgO particles as shown in the same figure were observed in some locations. Such MgO particles with a size of 20–30 nm, are attributed to the oxidation of magnesium coating on AlBO whisker during heat treatment.

The preforms of untreated whisker, SiO_2 binder added whisker, Mg-deposited whisker and $MgAl_2O_4$ coated whisker were prepared, respectively. Their compressive strengths are shown in Fig. 4. For the preform without binder addition, the strength was very low. If 3 mass% SiO_2 was added into the preform, the strength was approximately ten times higher than that without binder addition. Magnesium could also be regarded as a binder since the strength of Mg-deposited AlBO preform

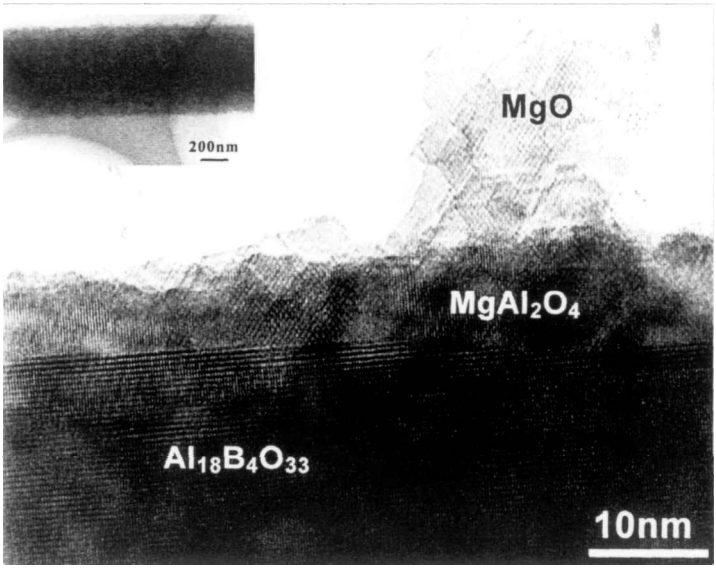

Figure 3. Surface profile HRTEM image showing the interface between AlBO whisker and $MgAl_2O_4$ coat (the top left one is a low magnification image).

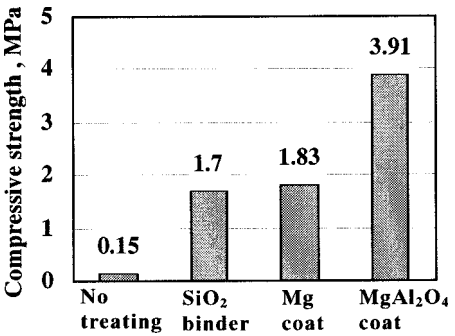

Figure 4. Compressive strength of four kinds of AlBO preforms.

was as high as that of the preform containing binder SiO_2. The preform having a $MgAl_2O_4$ coating was the strongest. SEM observation showed that the whiskers are connected with SiO_2 binder, Mg coat or $MgAl_2O_4$ coat (photographs are omitted), which was considered as the reason why those three preforms had a fairly high strength. Table 1 shows the change of the whisker volume fraction before and after casting. It can be seen that the stronger the preform was, the less were the changes of volume fraction and preform shrinkage. Therefore, Mg coating and $MgAl_2O_4$ coating created by the present process became good binders for the whisker preform.

From many studies it has been indicated that AlBO whisker reacts with magnesium included in the matrix during T6 treatment [5, 7, 10]. The whisker is damaged, and besides, the magnesium existing as the main element of age strength-

Table 1.
Change of whisker volume fraction in preform (composite) before and after squeeze casting

	Volume fraction (%)		Shrinkage ratio (%)
	Before casting	After casting	
No treating	19.0	24.0	20.8
SiO_2 binder	16.5	16.8	1.8
Mg coat	19.0	19.2	1.0
$MgAl_2O_4$ coat	19.0	19.0	0.0

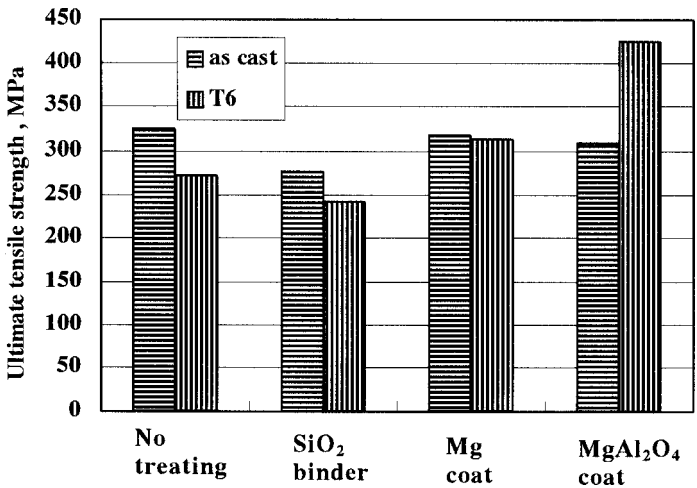

Figure 5. Tensile strength of AC4CH aluminum alloy composites reinforced by the four kinds of AlBO whiskers (Vf=20%).

ening phase (Mg_2Si) is exhausted. As a result, the composite strength after T6 treatment drops drastically. Figure 5 shows the ultimate tensile strength of the four kinds of whiskers reinforced AC4CH alloy composites in the as-cast and T6 states. Untreated AlBO/Al composite had about 16% strength deterioration after T6 treatment. Similarly, SiO_2 added AlBO/Al had about 13% strength loss in T6 state. This strength degradation contributes to the whisker damage and the consumption of magnesium in the matrix. However, in the case of Mg-deposited whisker, it may be judged that the whisker reacts just with this magnesium coating but not with the magnesium in matrix alloy. Although this reaction damages the whisker, the matrix composition does not occur and hence the age strengthening appears for the matrix alloy. Therefore, the composite strength degradation stopped after T6 treatment. Furthermore, in the case of $MgAl_2O_4$ coated AlBO/Al composites, because the reaction between the whisker and magnesium coating has been completed before squeeze casting, further whisker damage due to interfacial reaction can be avoided during T6 treatment. Therefore, composite strength in T6 state was considerably enhanced.

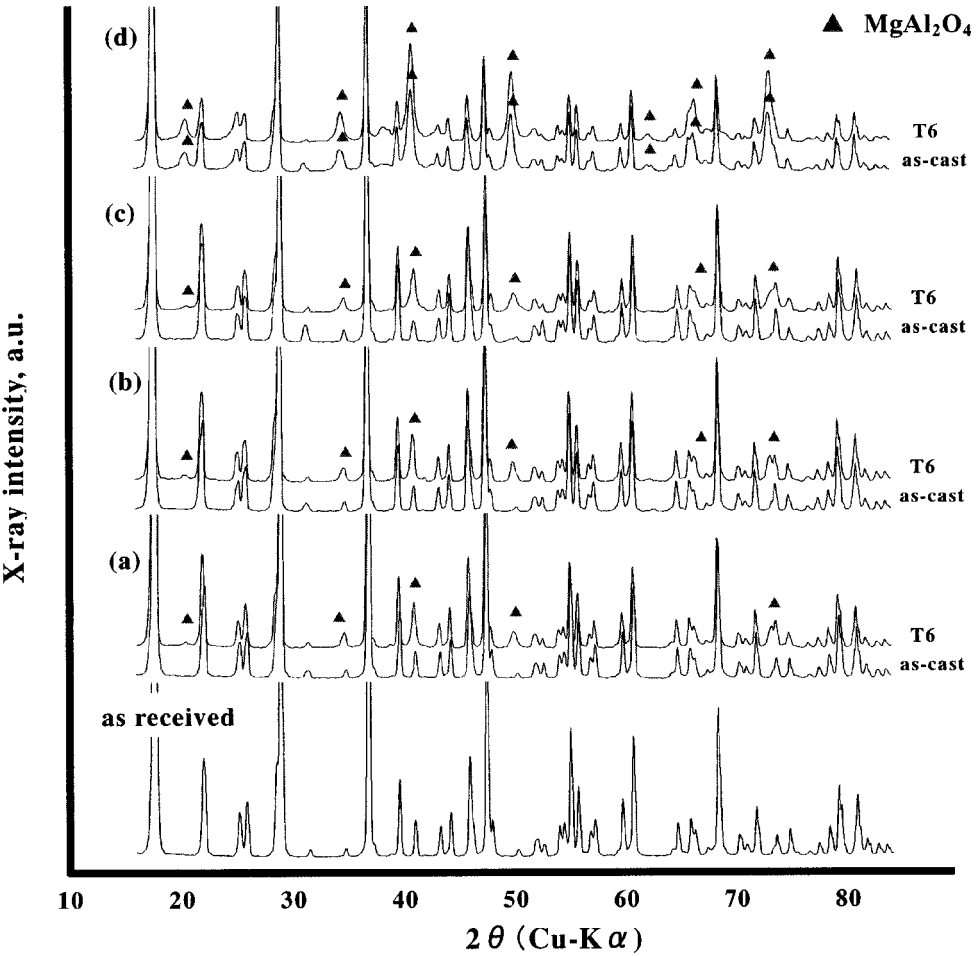

Figure 6. XRD patterns of the as received whiskers and the extracted whiskers from different AlBO/Al composites in the as-cast and T6 states. (a) Untreated AlBO, (b) SiO_2 added AlBO, (c) Mg coated AlBO, (d) $MgAl_2O_4$ coated AlBO.

Figure 6 shows XRD patterns of the whiskers extracted from the four kinds of AlBO/Al composites in the as-cast and T6 states. Figure 7 shows the corresponding SEM photographs of those whiskers. For the whiskers from the untreated and SiO_2 added AlBO reinforced composites in the as-cast state, although no $MgAl_2O_4$ was found by XRD analysis (Fig. 6a and 6b), SEM observation showed that many particle-like substances were covering the whisker. It means that an interfacial reaction occurred when the composites were fabricated. After T6 treatment, obvious $MgAl_2O_4$ peaks appeared in their XRD patterns. The whisker was nibbled by the further chemical reaction between AlBO and the matrix alloy (Fig. 7). The whisker surface became very rough and many large particle-like products were adhering to the whiskers. AlBO whisker damage by such an interfacial reaction

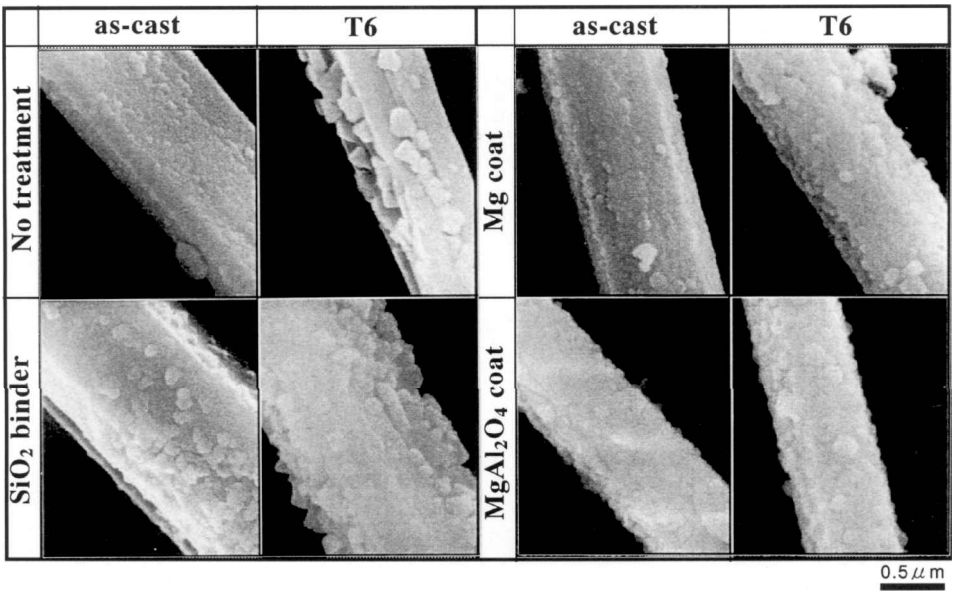

Figure 7. SEM photographs of the extracted whiskers from different treated AlBO whisker reinforced AC4CH alloy composites in the as-cast and T6 state.

resulted in drastic composite strength degradation. In the case of Mg coated AlBO whisker reinforced composite, XRD patterns of the whiskers from the as-cast and T6 states composites were similar to those of the untreated and SiO_2 added AlBO composites (Fig. 6c). A difference may be seen from SEM observation. The surface observation of the Mg-deposited whisker from T6 treated composite revealed further damage other than that of the as-cast composite, but the degree became much less compared to that of untreated and SiO_2 added AlBO composites (Fig. 7). The interfacial reaction was prevented partially and magnesium in the matrix alloy was not exhausted. This may be the reason why the strength of Mg coated AlBO composite did not decrease after T6 treatment.

Finally, in the XRD patterns of the extracted whiskers from $MgAl_2O_4$ coated AlBO composite, $MgAl_2O_4$ peaks appeared in the as-cast state (Fig. 6d). This is attributed to the chemical reaction between AlBO whisker and Mg coating during the treatment before squeeze casting. After T6 treatment, XRD peaks hardly changed. The surface state of AlBO whisker did not have any visible differences compared to that from the as-cast composite (Fig. 7). In other words, the whisker damage due to interfacial reaction has been stopped. The age strengthening effect in the matrix leads to the considerable increase in composite strength.

4. CONCLUSIONS

1. By a vacuum evaporation method, a uniform magnesium coating has been deposited on AlBO whiskers successfully. By the heat treatment of the Mg-

deposited whiskers at 800°C for 0.5 h in air, a continuous $MgAl_2O_4$ layer of 8–10 nm formed on the whiskers.

2. SiO_2, magnesium and $MgAl_2O_4$ coating can be regarded as the binders to strengthen the AlBO whisker preform. The deformation of preform using those binders was avoided when composites were fabricated.
3. Adding SiO_2 in the whisker preform did not change the interfacial reaction behavior and hence the strength of SiO_2 added AlBO/AC4CH alloy composite still decreased after T6 treatment.
4. Mg-deposited AlBO whisker could be kept from reacting with the matrix alloy during T6 treating, so the composite strength degradation was avoided.
5. $MgAl_2O_4$ coating on AlBO whiskers played the role of barrier to prevent the interfacial reaction. T6 treatment increased composite strength considerably.

REFERENCES

1. K. Suganuma, T. Fujita, N. Suzuki and K. Niihara, *J. Mater. Sci. Lett.* **9**, 633–635 (1990).
2. Shikoku Chemicals Industry Co. Ltd., *ALBOREX Technical Report*, 1992, pp. 2–4.
3. Y. Shintari, Y. Okochi and M. Sugiyama, *Proceedings of the 5th Japan International SAMPE Symposium*, 399–404 (1997).
4. T. Kitamura, K. Sakane, H. Wada, H. Hata and Y. Shintari, *Proceedings of International Workshop on Advanced Materials for Functional Manifestation of Frontier and Environmental Consciousness*, 11–18 (1997).
5. L. J. Yao, G. Sasaki and H. Fukunaga, *Mater. Sci. Eng. A* **225**, 59–68 (1997).
6. L. J. Yao, G. Sasaki, J. Pan, M. Yoshida and H. Fukunaga, *Metall. Mater. Trans. A* **29**, 1517–1524 (1998).
7. K. Suganuma, G. Sasaki, T. Fujita and N. Suzuki, *J. Japan Inst. Light Metals* **41** (5), 297–303 (1991).
8. K. Nagatomo and K. Suganuma, *J. Japan Inst. Metals* **58** (1), 78–84 (1994).
9. M. Shibata and Y. Takemoto, *J. Japan Inst. Metals* **64** (1), 1–6 (2000).
10. X. G. Ning, J. Pan, J. H. Li, K. Y. Hu, H. Q. Ye and H. Fukunaga, *J. Mater. Sci. Lett.* **12**, 1644–1647 (1993).
11. G. Sasaki, M. Yoshida, J. Pan and H. Fukunaga, *Mater. Sci. Res. Intern.* **5** (4), 276–279 (1999).
12. J. Pan, G. Sasaki, L. J. Yao and M. Yoshida and H. Fukunaga, *Mater. Sci. Technol.* **15** (9), 1044–1048 (1999).
13. X. G. Ning, J. Pan, K. Y. Hu and H. Q. Ye, *Materials Letters* **13**, 377–381 (1992).