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EFFECT OF WHISKER COATING TREATMENT ON THE STRENGTH OF $Al_{18}B_4O_{33}$ /AC4CH ALLOY COMPOSITES

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SUMMARY: By using a low vacuum evaporation technique under 10Pa at 650℃ for 1h, magnesium was deposited on $Al_{18}B_4O_{33}$ whiskers uniformly. After Mg-coated whisker preform was heat treated at 800℃ for 0.5h in air, $MgAl_2O_4$ layer with a thickness of 8-10nm grew on the whisker. The untreated, SiO_2 added, Mg-coated and $MgAl_2O_4$-coated whisker reinforced AC4CH aluminum alloy composites were fabricated by squeeze casting method. The composition and phase identification of the coatings were carried out with TEM, XRD, EPMA and SEM. The strength of whisker preform and composites were evaluated. The whisker preform having Mg or $MgAl_2O_4$ coating had higher strength, which eliminated the deformation of preform during squeeze casting. Those coatings could be regarded as whisker binder. When whisker was coated with Mg, interfacial reaction could be decreased because the composite strength did not drop down after T6 treatment. In the case of $MgAl_2O_4$-coated whisker, composite strength had a considerable enhancement. The reason was thought that $MgAl_2O_4$ had a closed bond to the whisker, which prevented the interfacial reaction of the whisker with aluminum matrix effectively.

KEYWORDS: Aluminum borate whisker, magnesium coating, spinel coating, aluminum alloy, squeeze casting, age treatment, tensile strength

INTRODUCTION

Aluminum borate ($Al_{18}B_4O_{33}$) whisker has drawn a considerable attention for the high cost-performance as a reinforcement of aluminum alloy or magnesium alloy, since it is commercialized in the beginning of 1990's. In comparison with the SiC whisker, $Al_{18}B_4O_{33}$ whisker reinforced aluminum alloy composites have excellent strength, elastic modulus, thermal expansion and wear resistance [1-2]. However, during fabrication and heat treatment of aluminum alloy composite material, the whisker reacts with the matrix easily, and leads to composite strength degradation. For instance, $Al_{18}B_4O_{33}$ reacts with magnesium including in aluminum alloy at about 520℃, and with aluminum directly at about 730℃ [3,4]. The heterogenous products like spinel ($MgAl_2O_4$), γ-Al_2O_3 are formed at the whisker surface [4-7]. This leads to deterioration in reinforcement efficiency owing to the whisker damage and whisker length shortening [8-10]. In addition, because of consumption of magnesium in the matrix alloy due to the reaction with whisker, the age strengthening of the matrix after T6 treatment cannot be obtained [10,11]. Therefore, a suitable whisker surface treatment to establish appropriate barrier layer seems necessary, in order to keep stable interface state of $Al_{18}B_4O_{33}$ whisker reinforced aluminum alloy composites. In the present work, a uniform magnesium coating is deposited on the whisker by vacuum evaporation of magnesium, and then a homogenous spinel ($MgAl_2O_4$) layer is created by the reaction between this magnesium coating and the whisker during heat treatment. The aim is to investigate the effect of different surface treatments on interfacial reaction and composite strength.

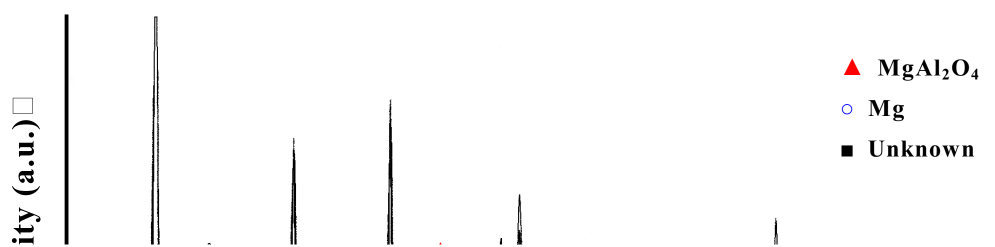
EXPERIMENT METHODS

$Al_{18}B_4O_{33}$ whisker, supplied 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 of whisker was carried out in a vacuum furnace. The whisker preform and a piece of pure magnesium were placed into the furnace firstly. Then they were heated under vacuum (10Pa). When the temperature arrived at 650℃, the vacuum valve was closed and the furnace chamber was kept in a sealed state. After heating at this temperature for 1h, electricity power was switched off, whisker preform was cooled down together with the furnace. The saturated evaporation pressure of magnesium at 650℃ is about 415Pa. It is expected that magnesium deposited on the whisker as a coating layer. Mg-deposited whisker was taken out from the vacuum furnace and then heated in atmosphere for 0.5h at 600℃, 700℃ and 800℃, respectively. The purpose is to form a spinel layer on the whisker by the reaction between the magnesium coating and the whisker. In order to compare the effect of surface treatment on composite characterization, 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℃, 760℃, 200℃ and 100MPa, respectively. The as-cast composites were T6 treated under the following conditions: solution treating at 525℃ for 8h, water quenching and aging at 160℃ for 6h. 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 the whiskers. Mg-deposited whiskers after heat treatment were observed using TEM (TOPCON/ EM-002B). The compressive strength of the whisker preforms and tensile strength of whisker/AC4CH composites were evaluated with a mechanical tester (SHIMADZU/ Autograph AG-100kNG).

RESULTS AND DISCUSSION

Characteristic of whisker surface coating

EPMA analysis result of Mg-deposited whisker showed that the atomic ratio Mg:Al was 13.5:86.5. From this result it can be confirmed at least that magnesium was coated on the whisker successfully. Since the analysis depth of EPMA is in micrometer order, detection of aluminum from the whisker implies that the thickness of magnesium deposition is very thin. XRD results of the whiskers after different treatments are exhibited in Fig.1. For the Mg-deposited whiskers, magnesium existence was hardly found (Fig.1b). This might be attributed to the state of magnesium. It is assumed to be a cluster without a



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perfect crystal. When the Mg-deposited whisker was heated at 600□, the peaks of magnesium and spinel were not identifiable (Fig.1c). But in the cases of heat treatment at 700°C and 800°C, obvious $MgAl_2O_4$ peaks could be confirmed (Fig.1d and 1e). For the whisker after heat-treatment at 800°C, the atomic ratio Mg:Al was 12.6:87.4, which was almost the same as that before heat-treatment. It is speculated that the reaction between magnesium and the whisker occurs as follow:

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

The calculation of Gibbs free energy ($\Delta G = -5.97\times10^6$ J/mol) indicated that this reaction can arise thermodynamically.

$Al_{18}B_4O_{33}$ whiskers that coated with magnesium at 650°C for 1h in vacuum and then heat-treated at 800°C for 0.5h in air were observed with transmission electron microscope. Fig.2 shows a surface profile image of a whisker. A continuous layer with a thickness of 8-10nm can be seen attaching on the whisker.

It adheres to the whisker keeping with a specific crystal orientation. The identification shows that it is a spinel phase $MgAl_2O_4$. This result is coincident with that of EPMA and XRD. Such a spinel layer bonded with the whisker closely should become a good barrier to prevent the reaction of the whisker with the matrix alloy. In the outside of the spinel layer, MgO particle as shown in the same figure was observed in some locations. Such MgO particle with a size of 20-30nm, are attributed to the oxidation of magnesium coating on the whisker during heat treatment in atmosphere.

Strength of whisker performs

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.3. 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

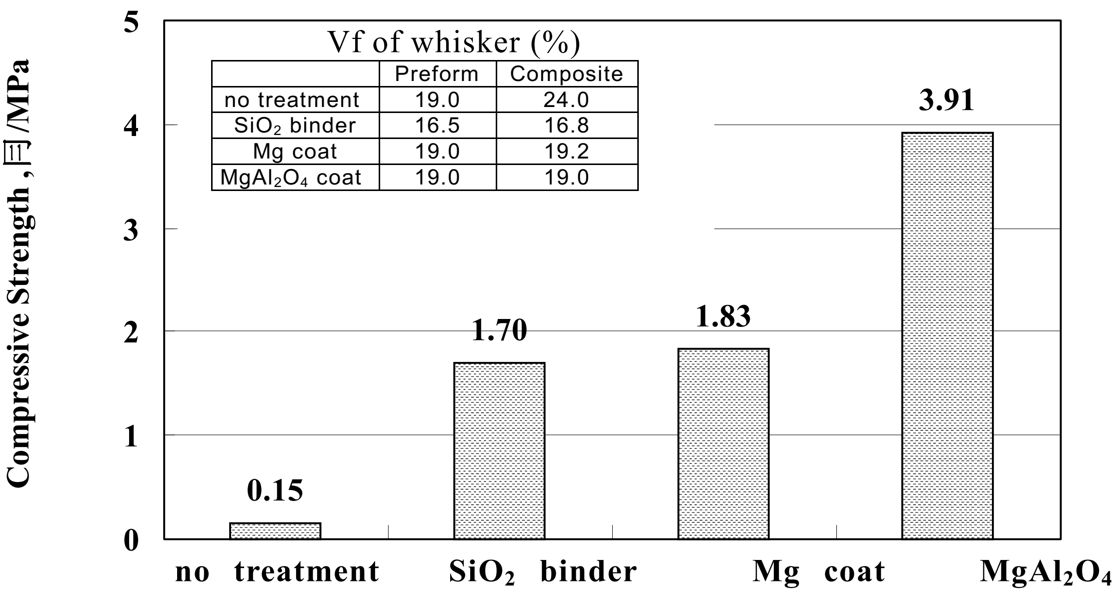
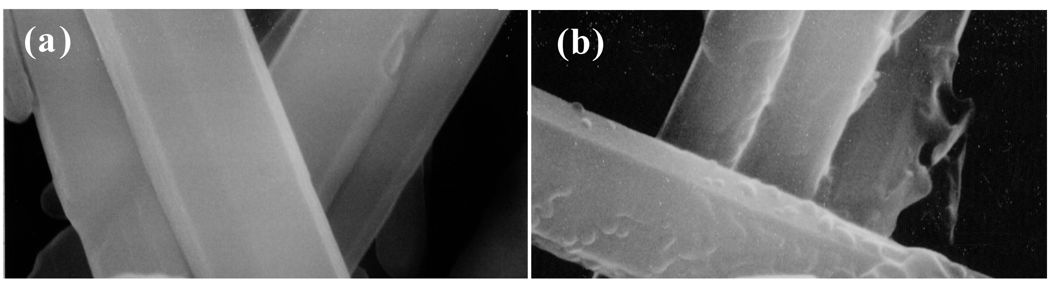


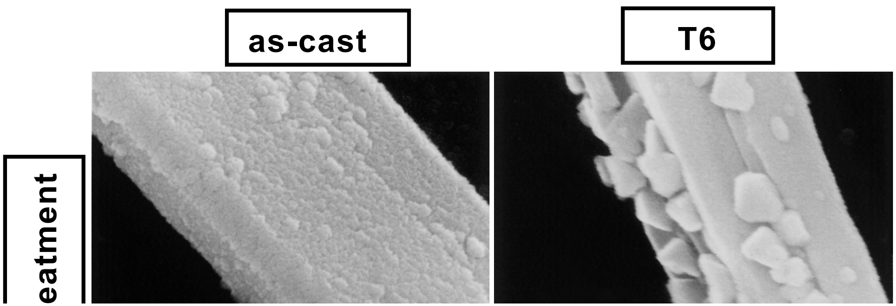
Fig.3 Compressive strength of four kinds of $Al_{18}B_4O_{33}$ preforms (whisker volume fractions in preform and composites are showed in the figure).



whisker preform was as high as that of the preform containing binder SiO_2. Furthermore, the preform having $MgAl_2O_4$ coating was the strongest. SEM observation shows that the whiskers are bridged with SiO_2 binder, Mg coat and $MgAl_2O_4$ coat (see Fig.4), which is considered as the reason why those three p reforms had a fairly high strength. Volume fraction change shows that the stronger the preform is, the less the changes of volume fraction and preform shrinkage is. Therefore, Mg coat and $MgAl_2O_4$ coat created by the present process are good binders for the whisker preform.

Effect of whisker surface treatment on composite strength

From many studies it is known that $Al_{18}B_4O_{33}$ whisker reacts with magnesium including in the matrix during T6 treatment [3,5,8]. The whisker is damaged, and besides, the magnesium existing as the main element of aging phase (Mg_2Si) is exhausted. As a result, the composite strength after T6 treatment drops


as-cast
T6
eatment

drastically. Fig.5 shows the ultimate tensile strength of the four kinds of whiskers reinforced AC4CH alloy composites and their matrices alloy in the as-cast and T6 states. Untreated whisker reinforced composite had 16% deterioration in strength after T6 treatment. Similarly, SiO_2 added whisker composite had about 13% strength loss in T6 state. This strength degradation is caused by 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 effect appears for the matrix alloy. Therefore, the composite strength degradation was stopped after T6 treatment. Furthermore, in the case of $MgAl_2O_4$ coated whisker/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 had a considerable enhancement. Fig.6 exhibits XRD spectra of the as-received whisker and the whiskers extracted from three kinds of composites in the as-cast and T6 states. Fig.7 shows the corresponding SEM photographs of those whiskers. For the whisker from the untreated whisker reinforced composites in the as-cast state, although $MgAl_2O_4$ peak was not found in XRD spectra (Fig.6b), SEM observation shows that many particlelike substances are covering the whisker surface (Fig.7a). It means that an interfacial reaction occurred when the composites were fabricated. After T6 treatment, obvious $MgAl_2O_4$ peaks appear in their XRD patterns. The whisker was nibbled by the further chemical reaction between whisker and the matrix alloy (Fig.7b). Whisker surface became very rough. Lots of large products are adhering to the whiskers. The whisker damage by such an interfacial reaction results in composite strength degradation drastically. In the case of Mg coated whisker reinforced composite, XRD patterns of the whiskers from the as-cast and T6 states composites are similar to that of the untreated whisker composites (Fig.6c). A difference may be seen from SEM observation. The surface observation of the Mg-deposited whisker from T6 treated composite reveals a further damage comparing to that of the as-cast composite, but the degree became much less than that of untreated and SiO_2 added whisker composites (Fig.7). It is assumed that the interfacial reaction was prevented partially and magnesium in the matrix alloy was not exhausted. It may be considered to be the reason why the strength of Mg coated whisker composite did not decrease after T6 treatment. Finally, in the XRD patterns of the extracted whiskers from $MgAl_2O_4$ coated whisker composite, $MgAl_2O_4$ peaks appeared in the as-cast state (Fig.6d). This is attributed to the chemical reaction between the whisker and Mg coating during the treatment before squeeze casting. After T6 treatment, XRD peaks almost did not change. The surface state of such whiskers did not have any visible differences comparing 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 composite strength increase considerably.

CONCLUSION

1. By a vacuum evaporation method, a uniform magnesium coating can be deposited on $Al_{18}B_4O_{33}$ whisker successfully. By the heat treatment of the Mg-deposited whisker at 800℃ for 0.5h in atmosphere, a continuous $MgAl_2O_4$ layer of 8-10nm can form on the whiskers.
2. Magnesium and $MgAl_2O_4$ coating can be regarded as a binder to strengthen the whisker preform. The deformation of whisker preform prepared by the present surface treatment can be avoided during fabrication of composites.
3. Adding SiO_2 in the whisker preform does not change the interfacial reaction behavior and hence the strength of SiO_2 added whisker/AC4CH alloy composite

drops down after T6 treatment.

4. Mg coating can decrease whisker damage due to interfacial reaction. After T6 treatment composite strength degradation is avoided.
5. $MgAl_2O_4$ coating plays a role of barrier to prevent the interfacial reaction. T6 treatment increases composite strength considerably.

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