SYNTHESIS OF TiC/Al COMPOSITE BY REACTIVE INFILTRATION TECHNIQUE

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SUMMARY: The spontaneous infiltration of the powder blend consisting of titanium and carbon with molten aluminum was investigated. The molten aluminum infiltrated into the [Ti + C] powder blend spontaneously, whereas the TiC powder phase was not infiltrated under the same experimental condition. Addition of aluminum powder in the [Ti + C] powder blend shortened the processing time and lowered the processing temperature to a large extent. The differental thermal analysis revealed that the aluminum powder in the powder blend melted at 933K and then ignited the reaction between titanium and carbon. The TiC formed *in situ* had a three-dimensionally continuous morphology. The optimum additional amount of aluminum in the powder blend was estimated to be in the range of 0.8-1.0 (Al/Ti molar ratio) .

KEYWORDS: aluminum matrix composite, infiltration, combustion reaction, titanium carbide,

INTRODUCTION

The infiltration of a ceramic preform with molten metals has been used as one of the processing routes for aluminum matrix composites [1,2]. Generally, non-reactive metal-ceramic systems have been used in this infiltration technique in order to prevent the degradation of the ceramic phase. Materials obtained by this process basically consist of a continuous metal matrix and discontinuously dispersed ceramic particles. Reactive systems have begun to be introduced into the infiltration process recently [3,4]. Since the ceramics are synthesized *in situ*, this process can provide a continuous ceramic matrix [5]. This is one of the outstanding features of this technique in that a bulk ceramic can be synthesized at relatively lower temperatures in a short time. Another advantage is that the highly reactive combination of a ceramic with a molten metal is in many cases wettable [6,7], which enables a spontaneous infiltration. In this work, titanium, carbon and aluminum powders were used as the starting materials, and the spontaneous infiltration of the powder blend with molten aluminum was attempted. As a result of the reaction between titanium and carbon shown below;

$$Ti + C \quad TiC \tag{1}$$

titanium carbide (TiC) is synthesized in molten aluminum. **Figure 1** shows a brief outline of the present work. The [Ti + C] powder blend is located at the bottom of a crucible, and an aluminum ingot is then located on the powder blend. The specimen is heated and as the temperature of the specimen increases, the molten aluminum penetrates into the spacing of the [Ti + C] powder blend. The TiC formation occurs in molten titanium subsequently.

The aim of this work is to synthesize TiC/Al composites via the infiltration route, and to this end the reactive infiltration of a compacted [Ti + C + Al] powder blend with molten aluminum is dealt with in this paper. We particularly focus on (i) observation of the composite microstructure and (ii) investigation of the effect of the aluminum addition on the reaction kinetics.

EXPERIMENTAL PROCEDURE

Infiltration process

The starting materials used in this work were titanium powder (99.8%Ti, with a particle size under 44μm), carbon powder (99.7%C, with a particle size of 5μm) and aluminum powder (99.8%Al, with a particle size under 45μm). The schematic illustration of the experimental setup for the spontaneous infiltration is shown in **Fig.2**. For the preparation of the starting material, the titanium and carbon powders were mixed with aluminum powder (Ti : C : Al molar ratio = 1:1:0-4.0). As is shown in Fig.2, the powder blend was placed at the bottom of the alumina (Al_2O_3) crucible (inner diameter: 13mm). An aluminum ingot was then placed on the powder blend. The chamber was evacuated using a rotary pump and backfilled with nitrogen gas (99.9% pure). The specimen was then heated up to a processing temperature in an induction furnace, and cooled in the furnace. The vertical cross-section was observed and analyzed by scanning electron microscopy (SEM), an electron probe X-ray microanalysis (EPMA) and X-ray diffraction (XRD) techniques.

Differential thermal analysis

Titanium, carbon and aluminum powders were mixed at molar ratios of 1:1 (Ti:C) and 1:1:1 (Ti:C:Al). The powder blends were consolidated by applying a pressure of 1000MPa and a piece of the compact was probed using differential thermal analysis (DTA). The analysis was conducted at a heating rate of 10K min^{-1}.

RESULTS

Advantage of using reactive system

As is described in the introduction, a chemical reaction was used to synthesize titanium carbide. To confirm the advantage of using the reactive infiltration technique, the infiltration behaviors of molten aluminum into (i) TiC powder and (ii) [Ti + C (Ti:C=1:1)] powder blend was compared. The result is shown in **Table 1**. The spontaneous infiltration of the [Ti + C] powder blend with molten aluminum occurred; whereas the TiC powder was not infiltrated by molten aluminum under the same experimental condition. **Figure 3** shows the temperature profile of the molten aluminum measured by a thermocouple inserted in the crucible during the heating process. The dotted line in the figure, which indicates the temperature profile of the specimen with the [Ti + C] powder blend, shows a relatively steeper increase in the temperature between points A and B. This indicates that an exothermic reaction took place during this period, which raised the temperature of the specimen. The reasons why using the [Ti + C] powder blend enables the spontaneous infiltration are considered to be twofold:

(a) The exothermic reaction raises the temperature of the whole system, which makes the wettability between aluminum and carbon better.
(b) Titanium powder becomes a path for molten aluminum’s infiltration.

Adding aluminum in the powder blends and the microstructure of the specimen

In the previous section, the spontaneous infiltration was confirmed by using the reactive system. However, a certain period of time was required to complete the reaction between titanium and carbon. **Table 2** shows the holding time and the processing temperature required for the complete TiC formation. Two specimens were prepared for this experiment (the [Ti + C (molar ratio, 1:1)] and the [Ti + C + Al (molar ratio, 1:1:1)] powder blends). As shown in **table 2**, adding aluminum in the powder blend lowered both the time and the temperature required for the TiC formation

Figure 4 shows the cross section of the specimen synthesized form the [Ti + C + Al] powder blend. A three-dimensionally continuous phase (brighter part in the figure) is clearly seen in an aluminum matrix (darker part). A qualitative analysis conducted by an XRD and EPMA revealed that the brighter phase in Fig.4 was TiC.

Role of aluminum in the powder blend

In order to confirm the role of aluminum in the powder blend, differential thermal analysis was carried out on both [Ti + C (molar ratio 1:1)] and [Ti + C + Al (molar ratio 1:1:1)] powder blends. The results of the DTA tests are shown in **Fig.5**. According to Fig.5, the [Ti + C + Al] powder blends showed an endothermic peaks (around 933K) followed by a sharp exothermic peak (TiC formation), whereas no clear peaks were found from the [Ti + C] powder blends. The role of aluminum in the powder blend is considered as follows;
(a) Aluminum melts at 933K.
(b) Molten aluminum starts to react with titanium (an exothermic reaction).
(c) The heat of reaction between Ti and Al ignites the reaction between Ti and C.
Thus the addition of aluminum decreases the ignition temperature of TiC formation to a melting point of aluminum.

Aiming at investigating the optimum additional ratio of the aluminum, six powder blends with different additional ratios of aluminum were prepared (Al/Ti molar ratio: 0.5-4.0). The compacted powder blends (diameter: 10mm, thickness: 10mm) were located on a heater and heated to induce the combustion reaction. **Figure 6** shows the temperature profile of the six powder blends. In this experiment, temperatures of the specimens were measured by a thermocouple embedded in each compacted powder. A large amount of heat, which could not be measured by the thermocouple, was released in cases where Al/Ti molar ratios were 0.8 and 1. In these specimens, the formation of TiC was observed after the combustion reaction; however, only relatively smaller increases in temperatures were detected when molar fraction of aluminum was 0.5, 0.7, 2.0 and 4.0. Thus, the optimum range of aluminum addition is roughly estimated to be in between 0.8 and 1.0.

CONCLUSIONS

The infiltration of molten aluminum with compacted [Ti + C] or [Ti + C + Al] powder blend was attempted in order to synthesize a TiC/Al composite; and the following results were obtained

1. The [Ti + C] powder blend was infiltrated with molten aluminum, whereas TiC powder was not infiltrated.
2. Adding aluminum powder in the [Ti + C] powder blend could lower the processing temperature and shorten the processing time. Aluminum in the [Ti + C + Al] powder blend react with titanium and increase the temperature of the specimen and ignite the reaction between titanium and carbon.
3. The optimum additional ratio of aluminum in the powder blend was estimated to be in between 0.8-1.0 (Al/Ti molar ratio).
4. The three-dimensionally continuous TiC was formed in the aluminum matrix as a result of the *in situ* reaction.

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Table 1 Possibility of spontaneous infiltration

Starting Materials	TiC Powder	[Ti + C] Powder
Possibility of Infiltration	Non-infiltrated	Infiltrated

Table 2 Holding time and processing temperature required for the complete TiC formation

Starting Materials	Processing Temperature	Holding Time
[Ti + C]	1473K	3600s
[Ti + C + Al]	933K	0s

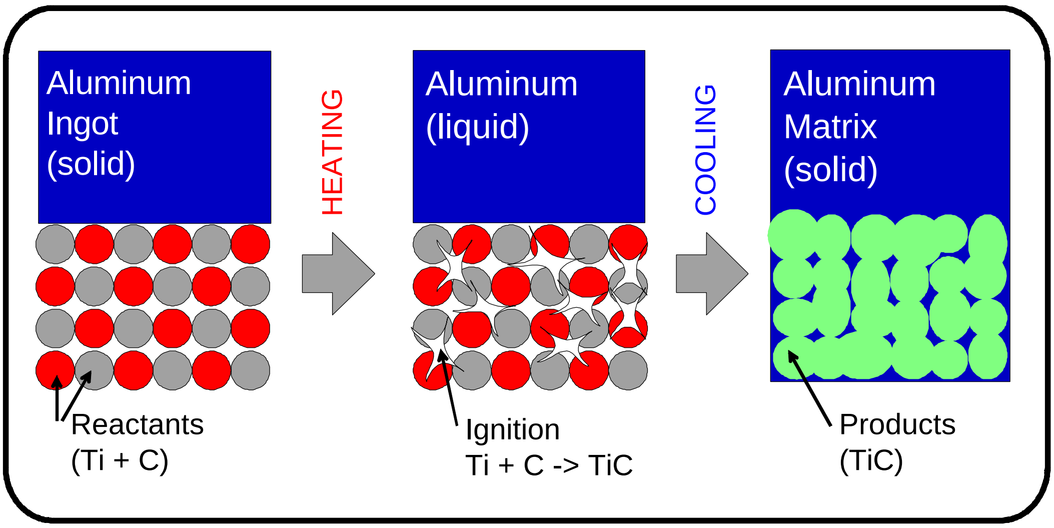

Fig. 1 Schematic illustration of the basic idea of the present work

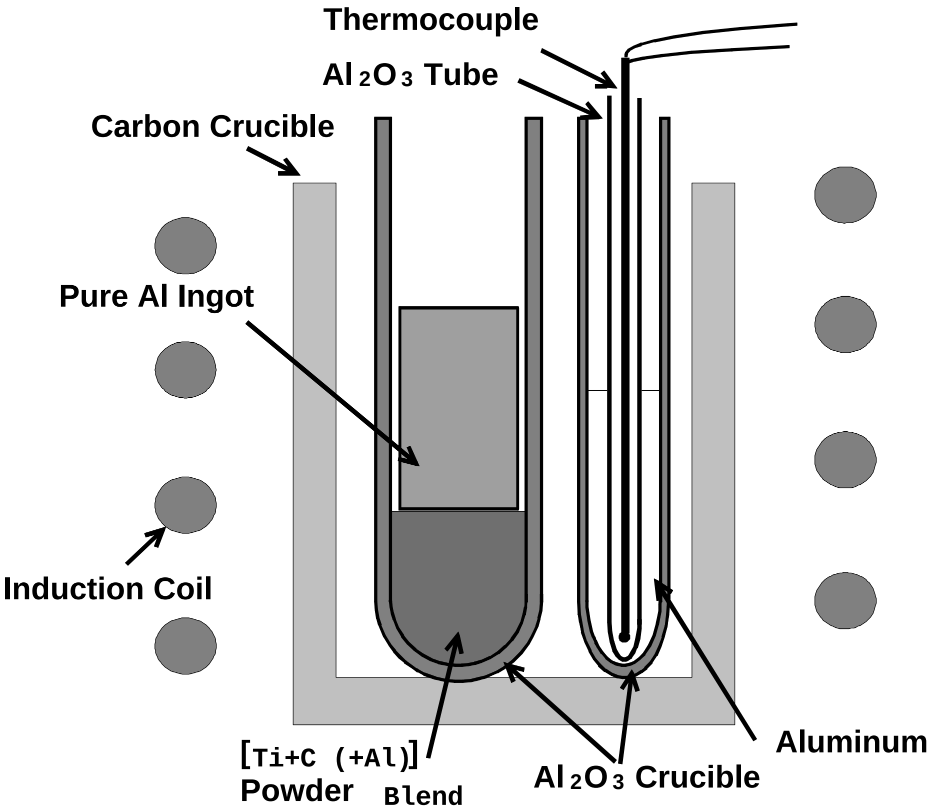

Fig. 2 Schematic illustration of the experimental set-up

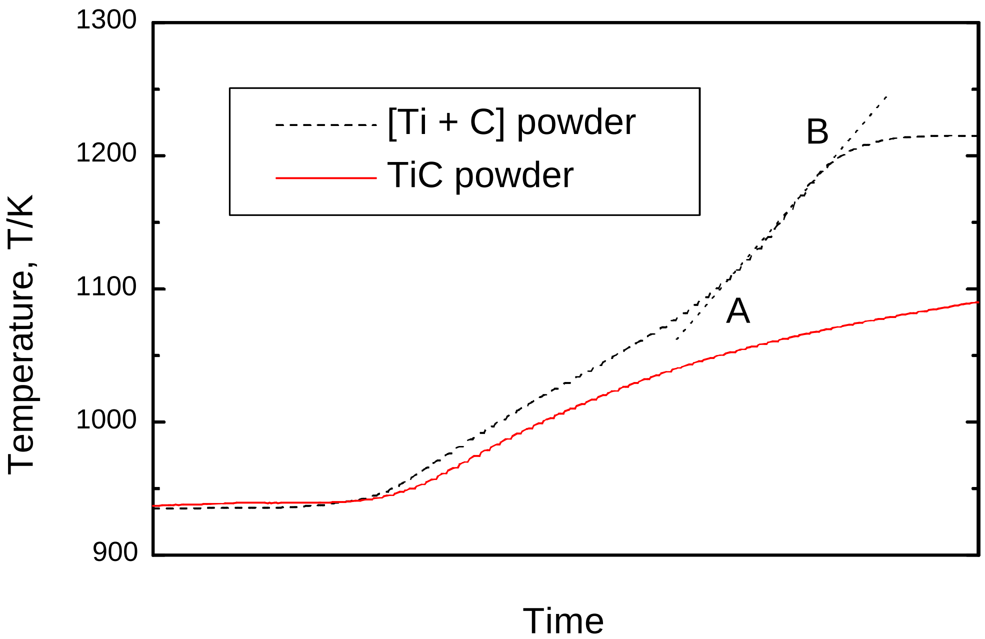

Fig. 3 Temperature profiles of molten aluminum during the heating process

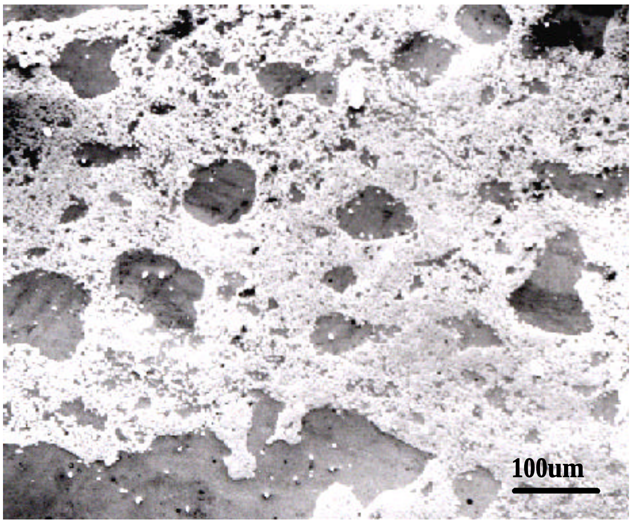

Fig. 4 Microstructure of the specimen made form the [Ti + C + Al] powder blend

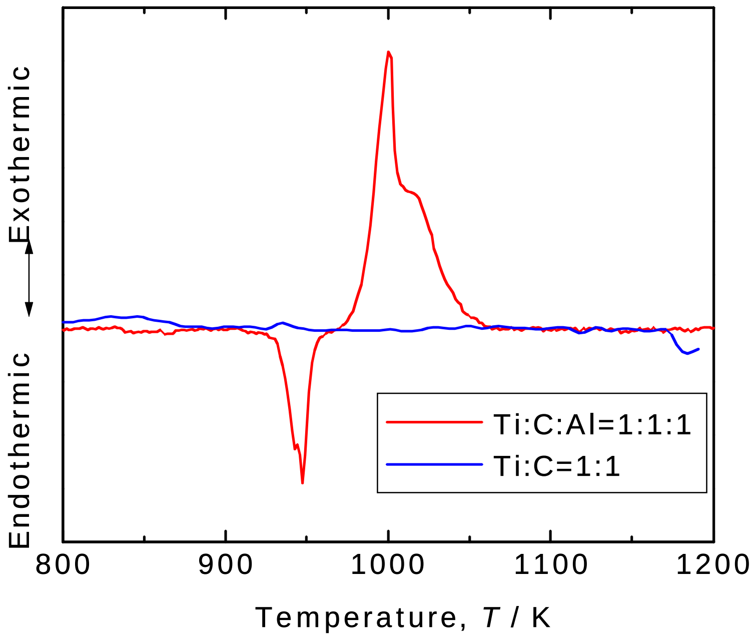

Fig. 5 DTA data carried out on the [Ti + C] and [Ti + C + Al] powders

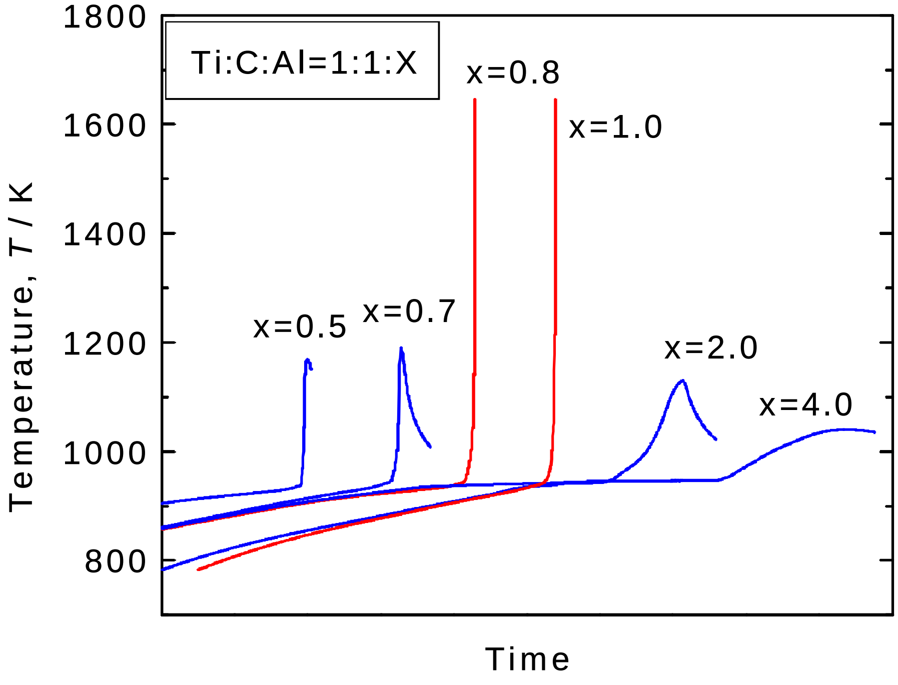

Fig. 6 Temperature profiles of the powder blends with various aluminum additional ratios