Properties of Ti-Aluminides-Reinforced Ti-Matrix Laminate Fabricated by Pulsed-Current Hot Pressing (PCHP)*

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Ti-aluminides-reinforced Ti-matrix composites were fabricated from 0.04 mm-thick Ti foils and 0.012 mm- and 0.024 mm-thick aluminum foils, in a process using a pulsed-current hot pressing (PCHP) equipment, and the effect of reaction temperature on properties of the composites was investigated. The composites were of laminated structure and composed of Ti and reacted layers containing Ti-aluminides. The composition of the reacted layers was dependent on the reaction temperature employed. Tensile testing at room temperature revealed that the reaction temperature was effective for the mechanical properties, including tensile strength, elongation and fracture mode, of the composites. The tensile strength and the elongation of composites fabricated at 1273 K from 0.04-mm-thick Ti and 0.012-mm-thick Al foils were 810 MPa and 3.64%, respectively, while they were 677 MPa and 3.44% for composites fabricated at 1173 K. Microstructure observations of fractured specimens showed that Ti layers of the composites fabricated at 1173 and 1273 K played a significant role in improving ductility by prohibiting the growth of numerous cracks emanating from Ti-aluminides.

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1. Introduction

Metal-matrix composites (MMCs) can possess a desirable combination of mechanical and physical properties, such as high melting points and low density. Such a unique combination makes MMCs attractive for use in various applications including compressor vanes and cylinder heads of automobiles engines and some applications in aircraft industries, where high strength is required at elevated temperatures. In any cases, however, the cost of materials is one important factor in determining whether they can be used in applications.1,2 Thus, it is important to develop new fabrication methods for making such MMCs in economic ways.

Combustion synthesis, also termed self-propagating high-temperature synthesis (SHS), is believed to be an environmentally benign technique and can be used in forming MMCs in-situ via reactions between constituent elements.3–5 This technique may also provide another feature, a relatively inexpensive process, because starting materials are metals in foil forms, which are available in a wide variety of thickness and purity, compared with specially pre-alloyed intermetallic foils. Ti-aluminides are considered to be attractive as reinforcement for a MMC, because of their high melting points, stiffness and low density. Ti-aluminides have, however, some disadvantages including brittleness at room temperature and a tendency for having a high density of cavities if produced by combustion synthesis without application of pressure. Therefore, we proposed a new processing technique to produce a laminate-type Ti matrix composite containing Ti-aluminides. The process is involved in pulsed-current sintering (PCS),6–10 combustion synthesis and vacuum hot pressing, and called a pulsed-current hot pressing (PCHP) process.11–13 Through this process, laminate-type Ti matrix composites with Ti-aluminides can directly be fabricated from foils of Al and Ti. In the present study, investigated is the effect of processing conditions on properties of Ti-aluminides-reinforced Ti-matrix composites synthesized by PCHP. Tensile properties of the composites are investigated on an Instron reinforcing machine and the results are presented along with microstructure observations and compositional analyses, which are carried out by means of scanning electron microscopy and EPMA techniques.

2. Experimental Procedures

Commercially available 99.5% purity Ti foils (0.04-mm thick) and 99.98% purity Al foils (0.012-mm or 0.024-mm thick) were alternatively stacked. The stacked foils (65 mm long, 20 mm wide, 0.7 mm thick) were placed in a cylindrical graphite die for a PCS machine (Sumitomo Coal Mining Co. Ltd., Model SCM DR. SINTER-1020) equipped with an electric power source and a pressure device.

Composites are fabricated in the following processing cycles.12–14 That is, a stack of foils is heated to a given reaction temperature at a heating ratio of 1.7 K/s under a vacuum of 2 Pa at a load of 2.7 kPa. In this stage of processing, Ti-matrix composites contain porous Ti-aluminides. Then, pressure loading is increased from 2.7 kPa to 32 MPa and held for 600 s, followed by furnace cooling after unloading. In this second stage of processing, a high electric pulsed-current is applied through carbon punches during pressing of the Ti and Ti-aluminides, causing spark discharge in gaps between Ti and Ti-aluminides and in pores in Ti-aluminides. The instant rise of high temperature by spark discharge causes evaporation and melting on the surface of Ti.
layers and pores, followed by bonding of Ti and Ti-aluminides and densification of them.

In the present study, two kinds of composites were synthesized by PCHP process at various reaction temperatures of 1173 K, 1273 K and 1373 K to find the effect of hot pressing temperature on material properties. Composites were produced from 0.04 mm-thick Ti foils and 0.012 mm-thick Al foils, and from 0.04 mm-thick Ti foils and 0.024 mm-thick Al foils. The former is hereafter called COMP12 and the latter COMP24.

After fabrication of the above-mentioned composites, some were sectioned and prepared for standard metallography using a scanning electron microscopy (SEM) equipped with compositional back-scattered electron imaging devices. The chemical composition of the phases present in the fabricated composite was determined by electron-probe microanalysis (EPMA) and X-ray diffraction techniques. The composites thus fabricated were machined to flat dog-bone-shaped tensile specimens, 65 mm long, 9 mm wide, 0.7 mm thick with a gauge length of 20 mm. Tensile tests were performed in air at 293 K on an Instron testing machine at a constant crosshead displacement rate of $8.3 \times 10^{-3}$ mm/s, giving an initial strain rate of $4.2 \times 10^{-4}$ s$^{-1}$. After tensile testing, some fracture surfaces of the tensile specimens were observed by scanning electron microscopy (SEM).

### 3. Results and Discussion

#### 3.1 Microstructure

Composites COMP12 and COMP24 fabricated by the PCHP process at various reaction temperatures were observed by SEM, and microstructures obtained are shown in Figs. 1 and 2. COMP12 fabricated at 1173 K and at 1273 K are seen to be composed of multi layers. These layers can be classified roughly into two kinds of areas, dark gray areas and light gray areas. In order to get some insight into compositional alternations at and near the dark grey areas in the composites, EPMA analyses were performed on 30 regions, indicated by numbers in Figs. 1(d, e, f) and 2(d, e, f), and the results obtained are tabulated in Tables 1 and 2, respectively.

As seen, Al foils are essentially alloyed entirely, leading to the formation of reaction zones seen as dark grey regions. As seen in Fig. 1 and Table 1, the reaction zones in COMP12 are composed of Al$_3$Ti (1), Al-rich Ti solid solution (2), TiAl (3), and Ti$_3$Al (4) layers when fabricated at 1173 K. When fabricated at 1273 K, on the other hand, reaction zones are composed of Al-rich Ti solid solution (7), TiAl (8) and Ti$_3$Al (9) layers. It should be noted that no Al$_3$Ti layer was formed when fabricated at 1273 K. In this case, however, Al-rich Ti solid solution and TiAl layers become larger in thickness and Ti layers also become larger in Al content, as compared with those obtained at 1173 K. When fabricated at 1373 K, on the other hand, instead of Ti-aluminides, a solid solution of Ti-21 at%Al alloy was formed. Formation of Ti-aluminides phases is more prominent at 1173 and 1273 K. Since in all cases the reaction temperatures used are far below both the solidus line of Al$_3$Ti (1623 K) and that of TiAl (1723 K), the observation described above indicates the uniqueness of the PCHP process with respect to the reaction behavior of constituent elements. It should be emphasized that the result obtained here indicates that plate-shaped Ti-Al alloy can be fabricated directly from Ti and Al.
Similarly to COMP12 described above, as seen in Fig. 2 and Table 2, the reaction zones in COMP24 are composed of Al$_3$Ti (1, 8), Al-rich Ti solid solution (3, 9), TiAl (4, 10) and Ti$_3$Al (5, 11) layers when fabricated at 1173 K or 1273 K. In these layers, Al$_3$Ti layers (1, 8) are composed of many particles and interfaces between the particles were clearly seen at higher magnification micrographs, Figure 4(d, e). Although this suggests weak bonding between the particles, bonding was slightly improved by increasing reaction temperature during PCHP. With increasing reaction temperature, Al$_3$Ti layers decrease and Al-rich Ti solid solution and TiAl layers increase in thickness. When fabricated at 1373 K, reaction zones in COMP24 are only of TiAl layer (14). Non-existence of Al$_3$Ti and Al-rich Ti solid solution layers in COMP24 fabricated at 1373 K may be caused by a similar mechanism described for COMP12 fabricated at 1373 K. Similarly to COMP12, Al content in Ti layer increased with increasing reaction temperature.

In all composites fabricated in this study, except COMP12 fabricated at 1373 K, Al content in Ti layers decreased with increasing the distance from the central part of the reacted zone. This compositional gradient may be responsible for the strong bonding between Ti and Ti-aluminides.

As described above (2. Experimental procedures), PCHP process is composed of two kinds of processing stages. At the first stage, Ti-aluminides are formed porously between Ti layers and at the second stage, bonding and densification of Ti and Ti-aluminides are carried out by heating them at a given reaction temperature for 600 s. In order to examine the effect of interdiffusion between Ti-aluminides at the second stage, changes in Al content in Al$_3$Ti layer by interdiffusion during PCHP were calculated based on Fick’s second law. Assuming direct bonding between Al$_3$Ti layer and TiAl layer without Al-rich Ti solid solution layer or that between Al$_3$Ti layer and Ti$_3$Al layer without Al-rich Ti solid solution layer, Al content in Al$_3$Ti layer in composites is expressed in the following equation.

$$
(C(x, t) - C_1)/(C_2 - C_1) = 1/2(\text{erfc}(x/2\sqrt{D_l}t))
$$

The initial and the boundary conditions can be described as follows:

![Fig. 2 Microstructures of COMP24 fabricated at various temperatures by PCHP process. (a, b, c) low magnification and (d, e, f) high magnifications at and near Ti-aluminides.](image-url)
Tensile stress-strain curves of COMP12 fabricated at various temperatures.

$$t = 0: \begin{align*}
    &x < 0, C = C_1 \quad (0.5 \text{ or } 0.25) \\
    &x > 0, C = C_2 \quad (0.75)
\end{align*}$$

and

$$t > 0: \quad \lim_{x \to +\infty} \left(\frac{dC}{dx}\right) = 0 \quad x = 0, C = (C_2 - C_1)/2 \quad (0.625 \text{ or } 0.5)$$

where $C(x, t)$ is Al content in Al$_3$Ti layer at distance $x$ from the surface after heating period $t$, $C_1$ the initial Al content of TiAl or Ti$_3$Al layer, $C_2$ the initial Al content of Al$_3$Ti layer, and $D_i$ the interdiffusion coefficient in Ti-aluminide $i$ ($i = \text{TiAl or Ti}_3\text{Al}$).

Interdiffusion coefficients in TiAl and Ti$_3$Al are reported by Sprengel et al.\textsuperscript{15,16} The thickness of an Al$_3$Ti layer in COMP24 fabricated at 1273 K is 3 µm and that in COMP24 fabricated at 1373 K is 10 µm, as shown in Figs. 2 and 3. Here we assume that Al content at the interface of the diffusion couple becomes $(C_2 - C_1)/2$ right after heat treatment starts and also assume that the Al content is constant during the heat treatment. Under these assumptions, Al content at the central area of the Al$_3$Ti layer after 600 s of heat treatment can be calculated by the eq. (1), using the above-mentioned interdiffusion coefficients and thicknesses. The results obtained are tabulated in Table 3. As seen in Table 3, Al content at the central area of Al$_3$Ti in COMP12 decreases to 74.84 and 74.68 at%, respectively, for diffusion couples of TiAl/Al$_3$Ti and Ti$_3$Al/Al$_3$Ti, after heating at 1273 K for 600 s. Al content in COMP24 is on the other hand, almost constant (~75.00 at%) after heating at 1373 K for 600 s. These calculated results indicate that the effect of interdiffusion for changing the crystal structure of Ti-aluminides from Al$_3$Ti to TiAl or Ti$_3$Al by heating for 600 s during PCHP is negligible. Hence, drastic structure change in the Ti-aluminides formed in composites during PCHP would be impossible if interdiffusion occurred, i.e., non-existence of Al$_3$Ti layer both in COMP12 fabricated at 1273 K and in COMP24 fabricated at 1373 K may be caused mainly by the melting, evaporation and condensation between the above-mentioned Ti-aluminide layers.

### 3.2 Mechanical properties

Tensile tests were carried out for composites COMP12 and COMP24 fabricated by PCHP and typical examples of stress-strain curves obtained at 293 K are shown in Figs. 3 and 4, respectively.

As seen in Fig. 3, COMP12 fabricated at 1173 K shows a tensile strength of 677 MPa and an elongation of 3.44%. A tensile strength and an elongation of COMP24 fabricated at 1273 K were 810 MPa and 3.64%, respectively. This increase in mechanical properties may be related to the solid solution

### Table 3 Calculated Al contents at the central area of Al$_3$Ti layer in composites after heating for 600 s at various temperatures.

<table>
<thead>
<tr>
<th>Interdiffusion couple</th>
<th>Al content at the central area of Al$_3$Ti layer $C$(at%)</th>
<th>Holding temperature $T$/K</th>
<th>Interdiffusivity of TiAl($D_{TiAl}$) or Ti$<em>3$Al($D</em>{Ti_3Al}$) $D$/m$^2$s$^{-1}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>COMP12</td>
<td>TiAl/Al$_3$Ti</td>
<td>0.7484</td>
<td>1276 $D_{TiAl} = 1.57 \times 10^{-16}$</td>
</tr>
<tr>
<td>COMP12</td>
<td>Ti$_3$Al/Al$_3$Ti</td>
<td>0.7468</td>
<td>1273 $D_{Ti_3Al} = 1.9 \times 10^{-16}$</td>
</tr>
<tr>
<td>COMP24</td>
<td>TiAl/Al$_3$Ti</td>
<td>0.7500</td>
<td>1371 $D_{TiAl} = 1.54 \times 10^{-15}$</td>
</tr>
<tr>
<td>COMP24</td>
<td>Ti$_3$Al/Al$_3$Ti</td>
<td>0.7500</td>
<td>1374 $D_{Ti_3Al} = 1.5 \times 10^{-15}$</td>
</tr>
</tbody>
</table>
hardening of the Ti matrix by the increase of Al content in Ti matrix (Fig. 3 and Table 2) and/or the improvement in ductility of Ti-aluminides due to non-existence of brittle Al\(_3\)Ti layers.

It should be pointed out that similar studies were also carried out by two research groups\(^{15,16}\) to investigate for the fabrication of composites consisting of Ti and Ti-aluminides using a vacuum hot pressing technique. As described above, the tensile strength (810 MPa) obtained in this study is higher than that (470 MPa) by Alman \textit{et al.}\(^{17}\) and that (550 MPa) by Enoki \textit{et al.}\(^{18}\) The elongation (3.64\%) obtained in the present study is also higher than that (1.8\%) by Enoki \textit{et al.}\(^{18}\) The difference in these mechanical properties may be caused by the following reasons. The main mechanism involved in PCHP is evaporation and condensation under pressure, while that in vacuum hot pressing is inter-diffusion of atoms. In the PCHP process, instant evaporation takes place by the passage of high current due to skin effects by spark discharge, followed by condensation. This evaporation cleans pore surfaces and boundaries between layers and/or between particles, and help metal foils and reaction areas bond together under compressive loading. Concurrently, the resultant composite is densified under compressive loading. When fabricated at 1373 K, the tensile strength and the elongation were 486 MPa and 0.4\%, respectively. The reason for the poor mechanical properties is not understandable at the moment. The elongation of COMP24 decreased with increasing reaction temperature during PCHP, while the tensile strength was almost unchanged regardless the reaction temperature employed. The decrease in the ductility of COMP24 with reaction temperature may be related to high content of Al in a solid solution of Ti in the vicinity of reacted zones, as shown in Fig. 4 and Table 3.

\subsection*{3.3 Fracture surface}

Tensile specimens of COMP12 and COMP24 were fractured in tension and the fracture surfaces were observed by SEM, and SEM micrographs obtained for fracture surfaces of COMP12 and COMP24 are shown in Figs. 5 and 6, respectively. As seen, both composites revealed similar fracture patterns. Typical, ductile dimples at Ti layers are seen along with numerous cracks at reaction zones for both composites fabricated at 1173 K and 1273 K. When the micrographs are viewed closely, it is found that the area fractured in brittle mode was wider in COMP24 than in COMP12. COMP12 and COMP24 fabricated at 1373 K, on the other hand, fractured in brittle mode and no ductile dimples were observed.

Some tensile specimens fractured were also sectioned parallel to the tensile direction for further examination of deformation characteristics of areas immediate beneath the fracture surfaces. SEM micrographs taken from cross sections of these composites are shown in Figs. 7 and 8. As seen, in the case of COMP12 fabricated at 1173 K, many cracks were formed in reacted zones. It should be noted that although many cracks were formed, they did not penetrate into Ti solid-solution layers. When fabricated at 1273 K, cracks in Ti-aluminides decreased in population, indicating improvement of bonding between Ti-aluminide layers. This improvement may be attributed to the non-existence of brittle Al\(_3\)Ti. A solid solution of Ti with high content of Al synthesized at 1373 K fractured in a brittle mode. From the
Fig. 6 SEM micrographs of fracture surfaces for COMP24 PCHP-processed at various temperatures, showing ductile dimple fracture modes at Ti layer and brittle fracture modes at Ti-aluminides. (a, b, c) low magnification. (d, e, f) high magnification.

Fig. 7 SEM micrographs in the vicinity of fracture surfaces for COMP12, fractured in tension, showing cracks in Ti-aluminides terminating on Ti layers. (a, b, c) low magnification. (d, e, f) high magnification.
observations described above, the best process parameter of reaction temperature for COMP12 is believed to be 1273 K. Similar deformation characteristics described for COMP12 were also observed for COMP24. In the case of COMP24 fabricated at 1173 K, many cracks were formed in reacted zones. When fabricated at 1273 K, cracks in Ti-aluminides decreased in population, indicating slight improvement of bonding between Al₃Ti particles. COMP24 fabricated at 1373 K fractured in a brittle manner and many cracks penetrating into Ti layers were formed in TiAl intermetallic layers. In order to offer desirable combinations of high stiffness and high toughness to Ti/Ti-aluminides composites, needed are not only strong bonding between Ti and Ti-aluminides but also high ductility of Ti matrix. In addition, the optimization of thickness ratios of Ti and Al foils may also be important in improving mechanical properties of composites made from Ti and Al foils. Pressure and hot pressing time during PCHP are also needed to tailor.

4. Summary

Ti-aluminides-reinforced Ti-matrix composites were fabricated from 0.04-mm-thick Ti foils and 0.012-mm- and 0.024-mm-thick aluminum foils, by a process using a pulsed-current hot pressing (PCHP) equipment, and the effect of processing condition on the properties of composites fabricated was investigated. The composites fabricated were of alternatively-laminated structure and composed of Ti and Ti-aluminides. The microstructure of the Ti-aluminides in the composites changed with increasing reaction temperature during PCHP. Tensile tests carried out for the composites at room temperature revealed that the elongation, strength and fracture mode of the composites were varied depending on reaction temperature during PCHP. The tensile strength and the elongation of the composite produced from 0.04-mm-thick Ti and 0.012-mm-thick Al foils at 1273 K attained to 810 MPa and 3.64%, respectively. Microstructure observations of fractured specimens showed that Ti layers of the composites fabricated at 1173 and 1273 K played a significant role in improving ductility by prohibiting the growth of numerous cracks emanating from Ti-aluminides.

REFERENCES


