Synthesis of MIL composites by various methods

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Abstract. Four methods such as thermal explosion, reactive sintering, reactive pressing, and explosive welding + sintering are considered for the obtaining of the Ti-TiAl\textsubscript{3} metal-intermetallic laminate composite. The microstructure and phase composition of the samples are studied by the X-ray diffraction, local X-ray spectrum and optical microscopy methods. The study shows that multi-laminate composites can be obtained by all four methods.

1. Introduction

Progress in the creation of new technological innovations mainly depends on the development and improvement of technologies for obtaining materials with required properties, so the creation of materials with desired structural and functional properties is currently an area of increased attention in materials science and technology. The new promising class of structural materials includes metal-intermetallic laminate (MIL) composite materials which are represented by a multilayer composition with alternating metal and intermetallic layers [1-5]. These composite materials are attractive for use in aerospace engineering and many other areas, and the methods for obtaining of MIL composite materials expand the functionality of laminate composites and the area of application.

This work considers four methods such as thermal explosion, reactive sintering, reactive compression, and explosive welding + sintering for the obtaining of Ti-TiAl\textsubscript{3} metal-intermetallic laminate composite materials. The microstructure and phase composition of the samples were studied by the X-ray diffraction, local X-ray spectrum and optical microscopy methods.

2. Materials and methods

0.3 mm thick titanium (VT1-0) and 0.15 mm thick aluminum (8011) foils, 0.5 and 0.6 mm thick titanium (VT1-0) and 1 mm thick aluminum (8011) plates, titanium (PTS) and aluminum (ASD-4) powders were used in the experiments.

To conduct synthesis in the thermal explosion mode (synthesis that occurs throughout the volume of the mixture), a powder mixture of titanium and aluminum with a stoichiometric composition of 37.2 wt\% Ti + 62.8 wt\% Al and 0.3 mm thick titanium foils were used to synthesize titanium tri-aluminide (TiAl\textsubscript{3}). The powder mixture was pressed into tablets with a diameter of 20 mm and a porosity of 10-15% (pore volume (%) to the total volume). A tungsten-rhenium 200 micron thermocouple placed in the powder layer of the sample was applied to determine temperature. The setup was specially designed to conduct the synthesis.
Ti and Al plates and a PM-12M muffle furnace were used for reactive sintering. Reactive sintering was conducted using external load to improve the contact between the foils. Before starting the experiment, the plates were subjected to abrasion and degreasing. The sample consisted of alternating titanium plates and 3 aluminum plates. The ratio of the number of titanium and aluminum plates was calculated considering the formation of TiAl$_3$ intermetallide and pure titanium layers during sintering. In general, the sample consisted of 10 titanium and 27 aluminum plates which were sintered at 700°C and 900°C for 2, 4 and 6 hours.

The reactive compression (reactive sintering together with constant pressure) was conducted using a special setup at the Institute of Metal Physics UB RAS (Yekaterinburg). In the work the Ti and Al plates were used, and temperature, pressure, and the process time were varied.

Explosive welding was conducted in an explosive chamber (Lavrentyev Institute of Hydrodynamics SB RAS, Novosibirsk). The samples consisted of 11 and 13 alternating titanium (0.5 and 0.6 mm in thickness) and aluminum (1 mm in thickness) plates with a size of 50×100 mm and of 21 titanium (0.6 mm in thickness) and aluminum (1 mm in thickness) plates with a size of 120×300 mm were obtained by explosion welding. The samples obtained by explosive welding were subjected to reactive sintering in a muffle furnace at the temperatures of 700 and 900°C for 2, 4, 6 and 8 hours.

The microstructure and phase composition of the samples were studied by the X-ray diffraction, local X-ray spectrum and optical microscopy methods.

3. Experimental results

3.1. Thermal explosion synthesis

Experiments with titanium and aluminum powders in the thermal explosion mode demonstrate that the powder mixture reacted completely with the formation of TiAl$_3$ intermetallide. Figure 1 shows the thermogram of the synthesis process in the thermal explosion mode. The average heating rate of the Ti/Al mixture at the initial stage was 1.27°C/s. When reaching the melting temperature of aluminum and during its melting, the temperature of the Ti/Al mixture continued to increase due to the heating of titanium, while the heating rate decreased to 0.42°C/s. The duration of this stage was about 50 seconds. Then, the temperature increased due to energy release during the synthesis of intermetallide. The critical temperature required to initiate a reaction was 700°C/s. There is a transitional period with duration of 25 s before the transition to the thermal explosion mode. The stage of thermal explosion proceeds at a temperature growth rate of 70°C/s for 16 s. The maximum temperature in the mixture (in the synthesis products) reaches 1320°C.

![Figure 1. Thermogram of the synthesis process in the thermal explosion mode.](image-url)
Figure 2 shows the structure of the multilaminate composite, obtained by the above method, consisting of titanium foil and intermetallide synthesized from a powder mixture (light layers-TiAl₃, dark layers-titanium foils). The samples are characterized by high porosity, low strength, as well as the problems of mechanical and physical and chemical compatibility of different materials at the boundary between the layers. The areas with increased porosity are formed at the contact boundary between the titanium layer and the synthesized intermetallide.

The experimental results show that the duration of the process has a significant effect. The synthesis of the Ti/Al stoichiometric mixture in the thermal explosion mode proceeds rapidly for 15–20 seconds. For such a short period of time, diffusion processes cannot fully develop. As a result, a strong bond between titanium foil and synthesized intermetallide is not formed.

3.2. Reactive sintering
During the sintering of the samples at T=700°C, after 2 hours, thin layers are formed between titanium and aluminum foils due to the diffusion processes. These layers provide a relatively strong bond between the foils, despite the fact that aluminum is not fully used to form intermetallide in this processing mode. At a sintering temperature of 900°C for 6 hours, a laminate sample consisting of alternating layers of titanium (light layers) and TiAl₃ intermetallide (figure 3a) was obtained. Two phases are identified in the x-ray spectrum of the sample: Ti and TiAl₃ (figure 3b). Nonuniform porosity is observed in the intermetallide layer with increasing the porosity (dark areas) in the center of the layer.

Figure 2. Structure of a laminate composite material obtained by the thermal explosion method.

Figure 3. Structure (a) and X-ray diffraction pattern (b) of the laminate composite after reactive sintering at 900°C for 6 hours (1-TiAl₃, 2-Ti).
3.3. Reactive pressing
Figure 4 shows the structure of the sample obtained by reactive sintering combined with pressing from titanium and aluminum foils at a temperature of 700°C. There is a strong contact between the layers of titanium and intermetallic. The pores are nonuniformly distributed over the area of the sample, and the porosity of intermetallic is much lower than that of the samples obtained by the methods described above. The XRF data show that the laminate composite consists of two phases: titanium and titanium tri-aluminide.

![Figure 4](image.jpg)

Figure 4. Structure of the layers of the sample after reactive pressing.

3.4. Explosive welding + sintering
In 2010, under the project RFBR (No. 10-08-02101-e_k) a method was proposed and implemented [6] for obtaining a metal-intermetallic laminate composite materials according to the following scheme: explosion welding of a multilaminate sample consisting of titanium and aluminum plates with further sintering in a muffle furnace, varying time and temperature parameters and excluding the pressure from the control parameters [7–12].

Figure 5a shows the structure of the composite after explosive welding, where the light layers are Al, the darker layers are Ti. Figure 5b demonstrates the structure of the composite after reactive sintering. A well-formed laminate structure with a strong contact between the layers of titanium and aluminum is formed after explosive welding. Wavy boundaries which are typical for such experiments are observed at the contact of the plates. After sintering, the structure of the composite material is as follows. The aluminum layer interacts with titanium with the formation of an intermetallic layer. The increase in the sintering time leads to the decrease in the thickness of the aluminum layers prior to their complete conversion into the intermetallic layer and to the decrease in the thicknesses of the titanium layers. The samples after sintering consist of alternating titanium layers and TiAl3 titanium tri-aluminide layers. The wavy form of the boundaries is preserved during the sintering process.

As the sintering time increases at a temperature of 700°C, the thickness of the titanium layers decreases due to the consumption of material for the formation of intermetallic layers. The thickness of the titanium layer as a function of the time of reactive sintering is shown in figure 6. Synthesis of the TiAl3 intermetallic layer proceeds at a much higher rate for the first 2 hours of sintering. According to the data of X-ray microanalysis, all the test samples consist only of titanium and TiAl3 intermetallic after 8 hours of sintering.

A characteristic feature of the synthesis of the intermetallic layer during reactive sintering without pressing is the formation of a layer with increased porosity in the center of the synthesized layer, as illustrated in figure 3a and, especially in figure 5b. The increase in porosity is caused by the inequality of the diffusion coefficients according to the Frenkel effect [13], when pores are formed near the contact boundaries of two solids due to the appearance of excess vacancies in one of them caused by the inequality of the coefficient of their mutual diffusion.
Figure 5. Laminate structure of the composite after explosive welding (a) and further sintering at 700°C for 8 hours (b).

Figure 6. Thickness of the titanium layer as a function of the time of sintering.

This fact eliminates the effect of preliminary explosive welding, although the pressure required to prevent the formation of pores in this case may be lower than without explosive welding. An additional advantage of this method for obtaining MIL composites is the reduction of the synthesis time in comparison with the method of reactive pressing.

5. Conclusion

Four methods such as thermal explosion, reactive sintering, reactive pressing, and explosive welding + sintering were considered for the obtaining of the Ti-TiAl₃ metal-intermetallide laminate composite. The study showed that the multilayered composite could be obtained by all four methods.

However it should be noted that there are disadvantages in each of these methods. After synthesis in the thermal explosion mode, high porosity and low strength are observed in the intermetallic layers of the samples. There are also problems concerning mechanical and physical and chemical compatibility of different materials at the boundary between layers, which leads to the absence of a strong bond between the titanium foil and synthesized intermetallide. High porosity and low strength were observed in the intermetallic layer after reactive sintering, and increased porosity was formed in
the middle of the intermetallic layer. A central layer of increased porosity was also formed after explosive welding and sintering, which reduces strength characteristics of the composite. The method of reaction compression partially solves the problem of high porosity in the intermetallic layer, but nevertheless, there are pores nonuniformly located in the layer, which requires an additional improvement of modes for the synthesis of metal-intermetallic laminate composite materials.

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