MICROSTRUCTURE EVOLUTION OF LAMINATED ALUMINUM BRONZE - INTERMETALLICS COMPOSITE

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Abstract

Using aluminum bronze and titanium foils, CuAl10Fe3Mn2 - intermetallic phases laminated composites have been fabricated through reactive bonding at 875 °C in vacuum. Investigations were concerned with the structural transformations of a bronze - Ti couple boundary. Holding for a few minutes resulted in the formation of a thin layer at the interface. Prolongation of the heating time lead to reactions in the liquid state and completely disappearing of titanium layers. Thus, the final microstructure consisted of alternating layers of intermetallics and unreacted CuAl10Fe3Mn2 bronze. The microstructure was revealed in optical and scanning electron microscopy (SEM). The study exhibited the presence of different reaction products in the diffusion zone and their chemical compositions were determined by X-ray microprobe analysis. The occurrence of different intermetallic compounds such as TiCu, Ti_2Cu, τ_1 ($TiCu_2Al$) and τ_2 (TiCuAl) was predicted from the ternary phase diagram Ti-Cu-Al. The predominant part of the intermetallic layers was the mixture of TiCuAl and $TiCu_2Al$ phases, since a liquid front of reaction was moving into the aluminum bronze. The microhardness of the reaction products and the elemental components was comparatively measured.

Keywords: Titanium, aluminum bronze, intermetallics, laminated composite, microstructure

1. INTRODUCTION

Since over two decades there has been significant interest in the fabrication and investigation of mechanical behavior of a variety of laminated metal composites, such as metal-metal [1], metal-ceramic [2] and metal-intermetallic [3-12] systems. Very interesting are metal-intermetallic laminated (MIL) composites that have the potential to perform various functions, such as blast mitigation, ballistic protection, heat exchange, thermal management and vibration damping [5]. MIL composites combine the good ductility and toughness of metals with the higher elastic modulus, higher strength, and lower density of intermetallics. Lamination improves many properties including fracture toughness, fatigue behavior, wear, corrosion and damping capacity. It also provide enhanced formability or ductility for brittle intermetallics [1]. There are two groups of production techniques of MIL composites: deposition or bonding. Sputter or vapor deposition techniques involve atomic scale transport of the component materials. Such nano-laminated materials are typically fabricated by depositing hundreds of alternate nanoscale layers and they have received significant interest due to their extremely high strength [12]. Deposition techniques require sophisticated manufacturing equipment and are too slow for practical applications. These factors increase the cost of component production and make these composites economically unattractive. Bonding techniques, e.g. diffusion bonding, transient liquid phase bonding and reaction bonding between metal foils have some advantages. They involve relatively simple processing and the size and the number of layers that can be produced are not limited [3]. Moreover, the laminated structure of the composite allows for variations in the layer thickness and phase volume fractions of the components simply through the selection of initial foils thickness. A great number of laminated composites have been produced using Al and Ni [3,6,8], Ti [5,10,11], Nb [4] or Fe [7] foils. Previous works also revealed that titanium-intermetallic and copper-intermetallic composites can be produced by reaction that occurs at the interface of Ti and Cu [8,10]. Aluminum bronzes are corrosion-resistant alloys of copper containing from 4 to 15 % aluminum and small amounts of other metals, used to make many machine parts and tools. Alloys containing approximately 10 % aluminum are fabricated by sand casting and gravity diecasting into strong objects, including ship propellers [13]. The laminated composites with aluminum bronze matrix could be considered for structural applications because of their lower density than monolithic bronze. This kind of laminated composites has been investigated by Tsai et al. [14]. In the present study, the reaction synthesis was employed to fabricate laminated composites in vacuum using aluminum bronze and Ti foils. The primary purpose of this study was to recognize the effect of high temperature on the structure and development of the interfacial zone between aluminum bronze and titanium. In order to do it, microstructural investigations of the reaction zone were carefully performed. After that formed intermetallic phases were identified and the progress of synthesis process with prolonged time was investigated. As a result, the laminated aluminum bronze-intermetallic composites with alternately located layers were produced and presented.

2. EXPERIMENTAL PROCEDURE

In the work, 0.1 mm thick foils of titanium and 0.3 mm thick foils of CuAl10Fe3Mn2 aluminum bronze were used to produce laminated bronze-intermetallic composites with controlled treating time, temperature and pressure. Chemical compositions and room-temperature mechanical properties of base materials are given in **Table 1** (UTS is ultimate tensile strength).

Table 1 Chemical compositions and mechanical properties of the base materials

Materials	Composition of elements (at.%)											
	Ti	Fe	C	Al	Cu	Mn	Ni	Zn	O	Sn	V+Cr	N
Titanium	99.51	0.09	0.08	0.07	-	-	-	-	0.18	-	0.05	0.02
CuAl10Fe3Mn2	-	2.78	-	10.75	83.36	1.98	0.76	0.29	-	0.08	-	-

	Yield strength (MPa)	UTS (MPa)	Elongation (%)
Titanium	250	320	24
CuAl10Fe3Mn2	325	580	12

Aluminum bronze and titanium foils were cut into 10 mm x 10 mm square pieces. The joining surfaces were polished on 1200 grade abrasive paper just before bonding. Any contamination on the surfaces was removed with cotton swabs in water. After that foils were rinsed in water and then in ethanol. After drying rapidly, they were stacked into laminates in an alternating sequence. There were used 5 pieces of aluminum bronze and 4 pieces of Ti. A pressure of 5 MPa was employed at room temperature in a specially constructed vacuum furnace to ensure good contact between the metals. Series of attempts allowed to find that a temperature of at least 870 °C was necessary for the start and rapid development of structural processes at the interface between aluminum bronze and titanium. The temperature was increased from 20 to 800 °C at a heating rate of 0.25 °C/s. The samples were heated in vacuum of 0.01 Pa at 800 °C for 1h under applied 5 MPa pressure to allow diffusion bonding of the layers. After that the foils were heated to 875 °C and held at this temperature from 5 to 60 minutes. The pressure was removed during this processing sequence with the purpose of eliminating possible expulsion of liquid phases. The temperature was then decreased slowly (cooling rate of 0.16 °C/s) to 700 °C and the pressure of 5 MPa was applied again. Finally, the samples were furnace-cooled to room temperature (**Figure 1**). After fabrication, the samples were cut using diamond blade and polished applying standard metallographic techniques. Microstructural observations were performed using a JEOL JMS 5400 scanning electron microscope and a Nikon ECLIPSE MA 200 optical microscope. The chemical composition of the phases was determined by an energy dispersive spectroscopy utilizing an ISIS 300 Oxford Instruments.

Before the samples were examined with the optical microscope they had been etched to reveal grain boundaries and the structure of the intermetallic layers. Vickers (HV0.1) measurements were performed by Matsuzawa microhardness tester.

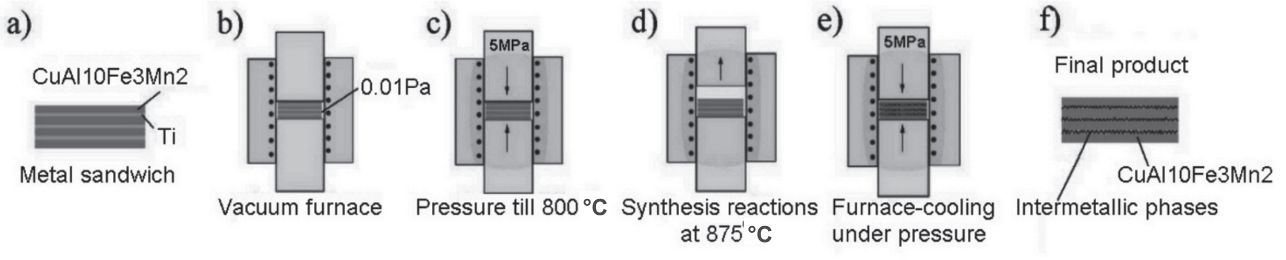

Figure 1 A schematic diagram of fabrication of CuAl10Fe3Mn2-intermetallic phases composite

3. RESULTS AND DISCUSSION

3.1. Structural transformations at the CuAl10Fe3Mn2-titanium boundary during reaction

At the beginning of the structural investigations a microstructure developed due to solid state diffusion between bronze and titanium was studied. **Figure 2a** shows the microstructure of the diffusion-bonded joint formed in the sample after holding for 1 hour at 800 °C.

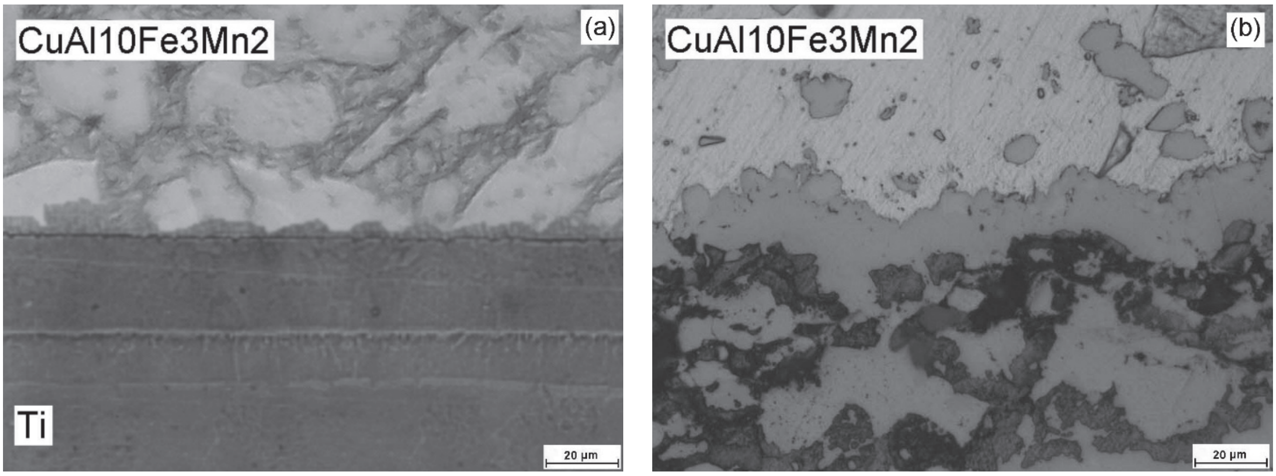

Figure 2 Optical micrographs showing the diffusion joint: (a) developed at 800 °C for 1 h and microstructure of the reaction zone, (b) formed after heat treatment at 875 °C for 30 minutes

In aluminum bronze-titanium diffusion zone four distinct reaction layers were observed. The analysis of the microstructure was based on the Cu-Ti-Al ternary phase diagram (**Figure 3**) using SEM and X-ray spectroscope. Adjacent to titanium the eutectoid mixture containing Ti_2Cu and αTi (solid solution of copper in titanium) were identified. This double-phase layer was followed by three single phase layers: TiCu (containing 49.6 at.% Ti, 48.8 at.% Cu, 0.94 at.% Al, 0.37 at.% Fe and 0.29 at.% Mn), TiCuAl (containing 32.61 at.% Ti, 37.11 at.% Cu, 29.85 at.% Al, 0.25 at.% Fe and 0.18 at.% Mn) and $TiCu_2Al$ (containing 24.94 at.% Ti, 49.11 at.% Cu, 25.11 at.% Al, 0.45 at.% Fe and 0.39 at.% Mn). **Figure 2b** shows the microstructure of the intermetallic layer formed after heat treatment at 875 °C for 30 minutes. The reactions with liquid transformed intermetallics to mushy stage and lead to completely disappearing of titanium layers. The measurements of the thickness of the reaction products showed that the reaction zone was increasing progressively with the reaction time. On the basis of measurements a simple relationship between the thickness (expressed in μm) of the intermetallics layer d and the holding time t (expressed in min) at the temperature of 875 °C was derived according to the **equation (1)**:

$$d = 0.03\, t^{2} + 45 \qquad (1)$$

The rate of synthesized layer growth with liquid phase contribution strongly exceeds the parabolic growth of intermetallic phases due to interdiffusion in the solid state.

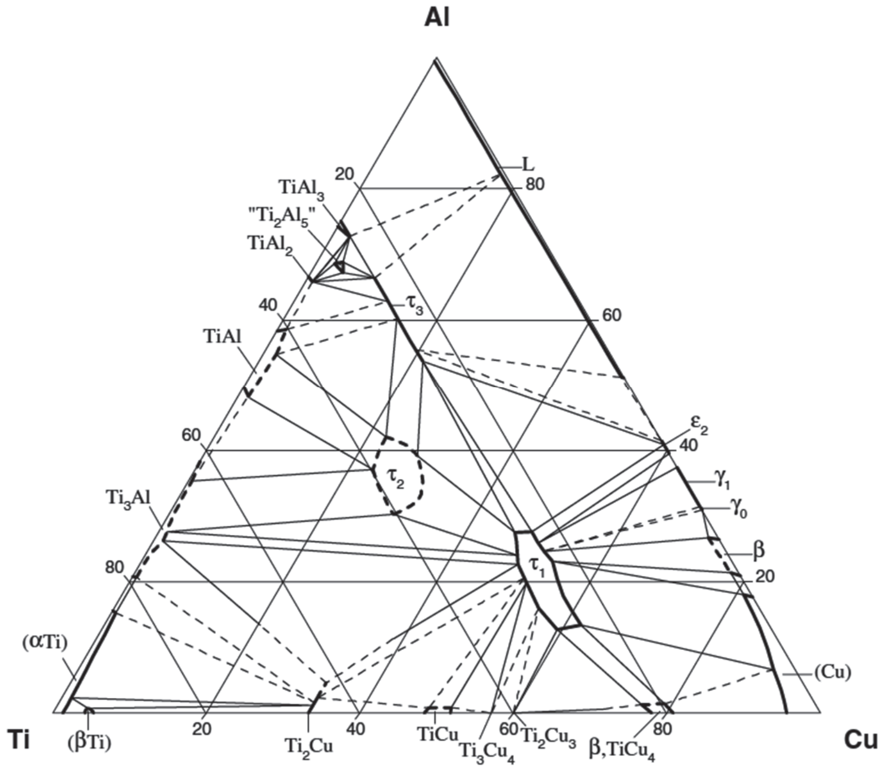

Figure 3 Isothermal cross-section through the ternary phase diagram Ti-Cu-Al at 800 °C [15]

The final microstructure consisted of alternating layers of intermetallics and unreacted CuAl10Fe3Mn2 bronze (**Figure 4**).

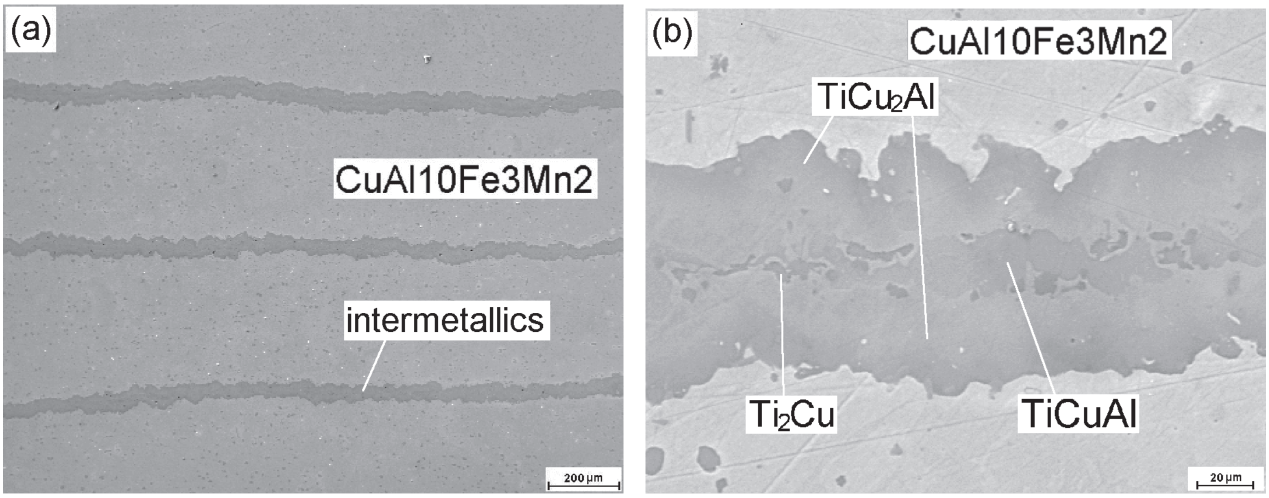

Figure 4 Scanning electron micrographs showing a typical microstructure of the laminated composite produced from aluminum bronze and titanium foils at 875 °C after treating for 60 minutes (a) and composition of the intermetallic layer in formed composite (b)

The X-ray analysis shown that intermetallic layers after treating for 60 minutes at the temperature of 875 °C were only composed of three intermetallics: $TiCu_2Al$ (containing 24.92 at.% Ti, 49.42 at.% Cu, 24.88 at.% Al, 0.41 at.% Fe and 0.37 at.% Mn), TiCuAl (containing 32.85 at.% Ti, 36.74 at.% Cu, 30.04 at.% Al, 0.23 at.% Fe and 0.14 at.% Mn) and Ti_2Cu (containing 61.91 at.% Ti, 31.01 at.% Cu, 6.17 at.% Al, 0.33 at.% Fe and 0.58 at.% Mn) that could dissolve up to 8 at.% Al (**Figure 5a**) [15]. The predominant part of the intermetallic layers were $TiCu_2Al$ and TiCuAl phases, since a liquid front of reaction was moving into the aluminum bronze (**Figure 5b**).

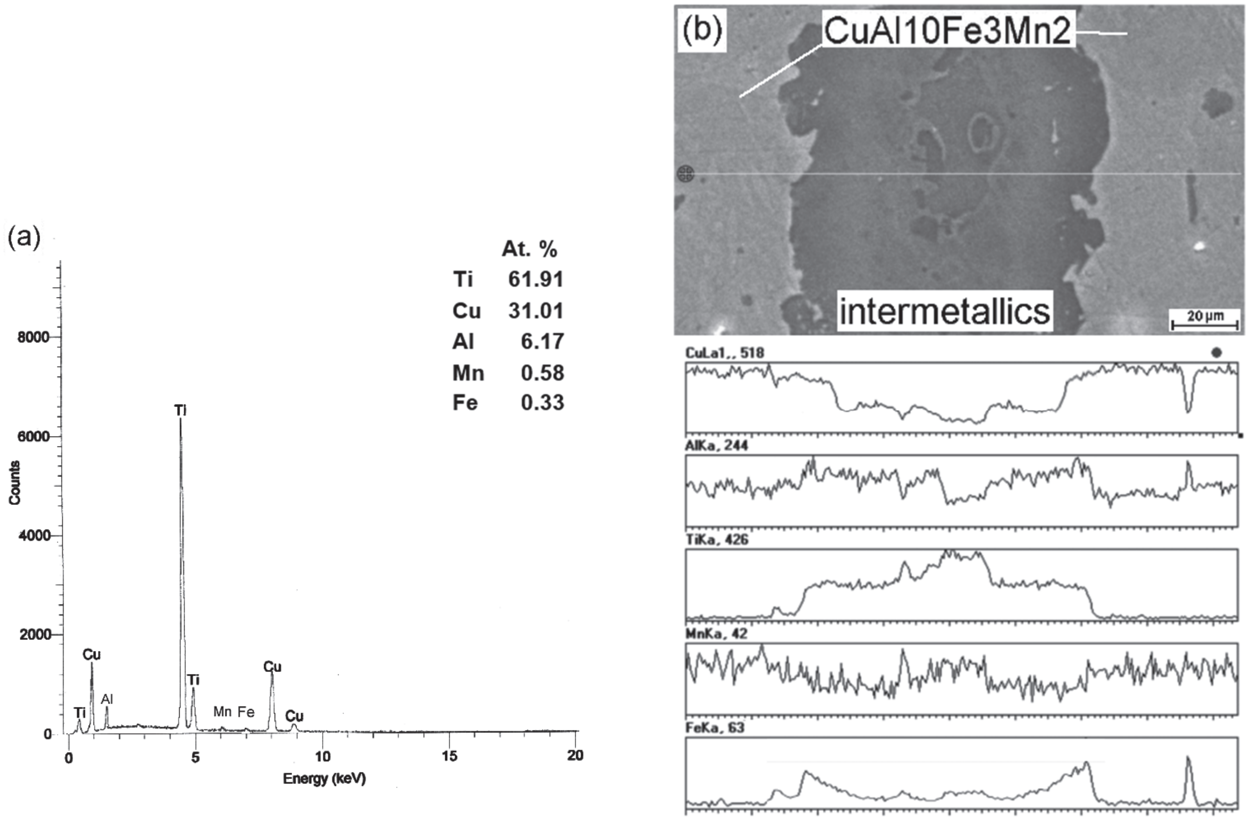

Figure 5 The energy spectrum for emitted X rays for Ti_2Cu phase (a) and concentration of Cu, Al, Ti, Mn and Fe profiles across the layers of the laminated composite (b)

3.2. Microhardness measurements

Microhardness measurements were performed for basic materials as well as for formed intermetallic layers. Results of microhardness measurements are given in **Table 2**.

Table 2 Results of hardness measurements

Material	CuAl10Fe3Mn2	Titanium	Bronze/intermetallic interface	Middle of the intermetallic layer
HV0.1	125-320	210-220	660-770	510-550

The maximum hardness values in the range of 660 to 770 HV were achieved at the bronze/intermetallic interface due to the presence of the $TiCu_2Al$ intermetallic phase. In the middle of the intermetallic layers, comprising mainly the mixture of TiCuAl and Ti_2Cu phases, hardness was a little bit lesser and contained in the range of 510 to 550 HV. In comparison, for materials used to produce the laminated composites: CuAl10Fe3Mn2 and titanium the values of hardness were from two to six times lesser.

4. CONCLUSION

In a consequence of reaction occurring between CuAl10Fe3Mn2 and titanium foils in vacuum at 875 °C, a laminated aluminum bronze-intermetallic composite can be formed. It is evident from metallographical examinations that the predominant part of intermetallics is synthesized in the region passing from a liquid state to a solid state. The reaction zone contains intermetallic compounds, presumably: Ti_2Cu, τ_1 ($TiCu_2Al$) and τ_2 (TiCuAl), but the predominant part of the intermetallic layers is the mixture of TiCuAl and $TiCu_2Al$ phases, since a liquid front of reaction is moving into the aluminum bronze. The intermetallic layers are from 2 to 6 times harder than layers of the aluminum bronze, and the maximum hardness values in the range of 660 to 770 HV can be achieved at the bronze/intermetallic interface due to the presence of the $TiCu_2Al$ intermetallic phase.

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