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TENSILE BEHAVIOUR OF LAMINATED TITANIUM-INTERMETALLIC COMPOSITE SYNTHESISED USING TITANIUM AND COPPER FOILS

Ti-intermetallic laminated composites have been fabricated *via* reaction synthesis in vacuum using 0.15, 0.20 and 0.25 mm thick foils of titanium and 0.05 mm thick foil of copper with controlled temperature and pressure. Effects of treating time at 900°C were studied by interrupting the reaction progressing after 0.5 and 5 hours. Microstructural investigations by scanning electron microscopy (SEM) and energy dispersive spectroscopy (EDX) showed that after 0.5 hour of heat treatment at 900°C intermetallic compounds: Ti_2Cu, TiCu, Ti_3Cu_4, Ti_2Cu_3, $TiCu_4$ were formed. The intermetallics layer consisted of thin layers of Ti_2Cu, thick layers of TiCu and the reaction zone consisting of $TiCu_4$ particles in Ti_3Cu_4+Ti_2Cu_3 matrix. Since titanium could diffuse through the Ti_2Cu and TiCu layers to the reaction zone, it leaded to the growth of TiCu at the expense of other phases. As a result, after 5 hours of treating the intermetallic layer was transformed almost wholly into TiCu, but with a thick Ti_2Cu interphase layer. The mechanical properties and fracture behaviour of the fabricated composites were examined through tensile test. The results showed that treating time at 900°C was a main factor determining properties because it led to an increase in volume fraction of the intermetallics. Unfortunately, long heat treatment caused a degradation of the intermetallic layers by oxidation because implemented vacuum was not high enough. After 0.5 hour of treating at 900°C the oxides on interlayer had no significant influence on the strength of the layers. EDX results showed that after long heat treatment the oxides were captured in the growing intermetallic layers to form inclusions or voids. This resulted in the formation of weak points in the intermetallic layers, from which cracks would have initiated easily, leading to premature failure of the layers during loading. Investigations indicated that the yield strength of all fabricated composites increased with increasing of the treating time. On the other hand, after 0.5 hours of treating the composites had higher ultimate tensile strength and higher strain at fracture. The results also showed that the composites exhibit a good cohesion between titanium layers and layers of intermetallic phases during tensile test.

Keywords: **laminated composite, intermetallics, mechanical properties, deformation**

ZACHOWANIE SIĘ PODCZAS ROZCIĄGANIA KOMPOZYTU WARSTWOWEGO TYTAN -FAZY MIĘDZYMETALICZNE WYTWORZONEGO Z FOLII TYTANOWEJ I MIEDZIANEJ

Używając folii tytanowych o grubościach: 0,15, 0,20 i 0,25 mm oraz folii miedzianej o grubości 0,05 mm, wytworzono na drodze reakcji syntezy kompozyt warstwowy tytan-fazy międzymetaliczne. Kontrolując temperaturę procesu oraz nacisk, doprowadzono do całkowitego przereagowania warstw miedzi, które z częścią tytanu utworzyły fazy międzymetaliczne. Zbadano wpływ czasu wygrzewania kompozytu w temperaturze 900°C na jego strukturę i własności. Reakcje syntezy faz przerywano po 0,5 i 5 godzinach. Badania z użyciem mikroskopu skaningowego i mikroanalizatora rentgenowskiego wykazały, że w kompozycie wygrzewanym przez 0,5 godziny występowały fazy: Ti_2Cu, TiCu, Ti_3Cu_4, Ti_2Cu_3,$TiCu_4$. Warstwy faz międzymetalicznych składały się z cienkich warstw fazy Ti_2Cu, grubych warstw fazy TiCu i obszarów zawierających cząstki fazy $TiCu_4$ w osnowie z mieszaniny faz Ti_3Cu_4+Ti_2Cu_3. Ponieważ tytan mógł dyfundować przez warstwy faz Ti_2Cu oraz TiCu, powodowało to wzrost ilości fazy TiCu kosztem pozostałych faz. Po 5 godzinach wygrzewania warstwy faz międzymetalicznych składały się prawie wyłącznie z fazy TiCu i cienkich warstw fazy Ti_2Cu. Przeprowadzono próby rozciągania uzyskanych kompozytów i stwierdzono, że głównym czynnikiem wpływającym na własności mechaniczne był czas ich wygrzewania w temperaturze 900°C. Wzrastał wówczas udział objętościowy faz międzymetalicznych oraz następowała degradacja warstw spowodowana ich utlenianiem. Badania z użyciem mikroanalizatora rentgenowskiego wykazały, że po 30 minutach wygrzewania tlenki występowały na granicy między tytanem i warstwami faz międzymetalicznych. Natomiast po 5 godzinach wygrzewania tlenki dostawały się do wnętrza warstw faz międzymetalicznych, powodując powstawanie inkluzji, które mogły być łatwymi zarodkami pęknięć. Dlatego kompozyty po 30 minutach wygrzewania charakteryzowały się większą wytrzymałością na rozciąganie oraz większym wydłużeniem. Z wyników prób rozciągania wynika także, że granica plastyczności wszystkich badanych kompozytów, niezależnie od grubości użytych do ich wytworzenia folii, wzrastała wraz ze wzrostem czasu wygrzewania w temperaturze 900°C. Badania wykazały również, że w trakcie próby rozciągania kompozyty wykazywały dobrą kohezję pomiędzy warstwami tytanu i faz międzymetalicznych.

Słowa kluczowe: **kompozyt warstwowy, fazy międzymetaliczne, własności wytrzymałościowe, deformacja**

INTRODUCTION

Metal-intermetallic composites offer an attractive combination of physical and mechanical properties distinct from the separate constituent phases, e.g. high toughness of the metal coupled with low density, high modulus and high strength of the intermetallic. Methods for the production of laminated composites, consisting of at least one brittle phase, include diffusion bonding [1], magnetron sputtering [2], vacuum plasma spraying [3] and synthesis reactions between dissimilar elemental metal foils [4-6]. Reaction synthesis of multilayers using foils has some advantages over some other methods. The obvious economic benefit in using this technique lies in the ease of processing. Furthermore, the process uses readily available elemental foils, which can easily be shaped prior to the initiation of the synthesis reaction, resulting in the potential to produce near-net-shaped composites. This technique has been recently implemented to produce Cu-intermetallic and Ti-intermetallic composites using Ti and Cu foils [7-9]. Previous works devoted to the mechanism of damage evolution and fracture behaviour of the metal-intermetallic laminates concern mainly the metal-aluminide [10-15] composites. The aim of this work was to study the tensile behaviour of the Ti-titanide laminated composites formed through *in-situ* reactions between Ti and Cu foils.

EXPERIMENTAL PROCEDURE

Commercially pure Ti (0.15, 0.20, 0.25 mm thick, 99.1 wt. %) and Cu (0.05 mm thick, 99.99 wt. %) foils were used to fabricate laminated titanium-intermetallic composites. Foils with dimensions of 50 mm x 10 mm were cut from the titanium and copper sheets and polished on 800 grade abrasive paper. After rinsing in water, rinsing in ethanol, and then drying rapidly, foils were laminated alternately into titanium/copper multilayer samples (11 Ti and 10 Cu layers) and then placed in a vacuum furnace. A pressure of 5 MPa was applied at room temperature to ensure good contact between foils. The temperature was initially raised to 850°C and then the pressure was released to avoid an expulsion of liquid phases. Next, the temperature was raised to 900°C, which was necessary for the start and rapid development of structural process at the interface of titanium and copper. The samples were held at a temperature of 900°C for 0.5 and 5 hours and then finally furnace-cooled to room temperature with cooling rate of 0.16°C/s. Vickers measurements were conducted using a Hanemann microhardness tester mounted on NEOPHOT 2 microscope under load of 0.637 N for 15 s. Samples with dimensions of 50 mm x 8 mm x 3 mm, made from fabricated composites, were subjected to tension test on an INSTRON screw machine at a constant crosshead speed of 0.1 mm/min. The crack propagation traces and fracture surfaces were examined using Nomarski contrast. Before the samples were examined with the optical microscope they had been etched using an aqueous HF solution (2%) to reveal any titanium grain boundaries and the structure of the intermetallic layers. Microstructural investigations were performed also using an scanning electron microscope JMS 5400. The chemical composition of the phases was determined by energy dispersive spectroscopy (EDX) using ISIS 300 Oxford Instruments.

RESULTS AND DISCUSSION

Detailed information concerning the structural changes during formation of laminated titanium-intermetallic composites has been published previously [8, 9]. Microstructural investigations showed that after 0.5 hour of heat treatment at 900°C the intermetallics layer consisted of thin layers of Ti_2Cu (adjacent to titanium layers), thick layers of TiCu and the zone consisted of $TiCu_4$ particles in Ti_3Cu_4+Ti_2Cu_3 matrix (Fig. 1). Since titanium can diffuse through the Ti_2Cu and TiCu layers to the reaction zone, it leads to the growth of TiCu at the expense of $TiCu_4$, Ti_3Cu_4 and Ti_2Cu_3. After 5 hours of treating the intermetallic layer was transformed almost wholly into TiCu, but with a thick Ti_2Cu interphase layer.

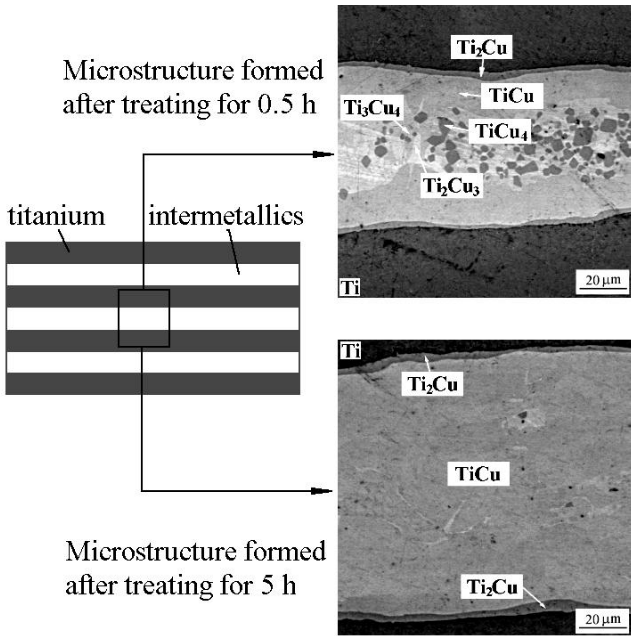

Fig. 1. Microstructures of the intermetallic layers in composites treated at 900°C for 0.5 and 5 hours

Rys. 1. Mikrostruktury warstw faz międzymetalicznych uzyskanych po 0,5 oraz 5 godzinach wygrzewania w temperaturze 900°C

According to Colinet et al. [16] the formation of TiCu as the dominant phase is thermodynamically favoured and can be understood from the steps involved in another phases formation, which occurs through a series of the solid-liquid and the solid-state reactions. Alman et al. [4] pointed out that the initial foil thickness has no effect on the composition of the formed intermetallic phases. Results show that the thickness of the elemental foils affects only the final thickness of the resultant layers. Intermetallic volume fraction V_i can be expressed as

$$V_i = \frac{r}{(1-w)\cdot\frac{\rho_i}{\rho_{Cu}} + (1-w\cdot\frac{\rho_i}{\rho_{Ti}})\cdot r} \tag{1}$$

where r is ratio of the original copper to titanium thickness, w is titanium weight percent in the intermetallic product, ρ_i is the density of intermetallic layers, ρ_{Cu} and ρ_{Ti} are the density of copper and titanium, respectively [17]. Table 1 summarises the microstructural features of the Ti-intermetallics composites.

TABLE 1. **Summary of microstructures of investigated composites**

TABELA 1. **Podsumowanie strukturalnych zależności w badanych kompozytach**

Time at 900°C, h	Starting Ti-foil thickness mm	Approximate composite layer thickness, mm		Volume fraction of titanides layer, %	Phases present
		Ti	Titanides		
0.5	0.15	0.129	0.071	47	TiCu $TiCu_4$ Ti_2Cu_3 Ti_3Cu_4 Ti_2Cu
	0.20	0.172	0.077	31	
	0.25	0.225	0.074	25	
5	0.15	0.084	0.116	58	TiCu Ti_2Cu
	0.20	0.127	0.122	49	
	0.25	0.181	0.118	39	

The results of the microhardness measurements for TiCu, Ti_2Cu, the mixture of phases ($TiCu_4$, Ti_2Cu_3 and Ti_3Cu_4) and Ti are 783, 681, 530 and 273 HV, respectively. The titanide phases (especially TiCu) give high hardness to the composite, while unreacted titanium provides ductility. Figure 2 shows the room-temperature tensile stress-strain curves for the laminated composites formed for 0.5 and 5 hours at 900°C.

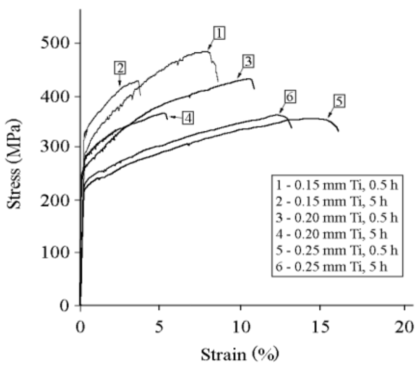

Fig. 2. Tensile stress-strain curves for the titanium-titanides laminated composites

Rys. 2. Krzywe rozciągania kompozytów warstwowych tytan-fazy międzymetaliczne

Table 2 presents the room-temperature tensile behaviour of these composites.

TABLE 2. **Tensile properties of Ti-titanides laminated composites**

TABELA 2. **Własności kompozytów warstwowych Ti-fazy międzymetaliczne uzyskane w próbie rozciągania**

Treating time at 900°C, h	Starting Ti-foil thickness mm	Yield strength MPa	Ultimate tensile strength MPa	Strain at fracture, %
0.5	0.15	286	474	8.4
	0.20	252	425	10.7
	0.25	218	352	16.2
5	0.15	328	421	3.8
	0.20	279	365	5.4
	0.25	226	359	12.9

The serrations in the stress-strain curves in Figure 2 in the region beyond the yield point correspond to the formation of multiple cracks in the intermetallic layers after application of the tensile load. The failure and structure evolution in the course of straining of this composite has been investigated and can be described in the following way. At the early stage of plastic straining titanium grains of the composite were deformed, whereas no traces of plastic deformation were noticed in the intermetallic layers. Formation of cracks in the layers of intermetallic phases was the characteristic feature of the prolonged composite deformation. The light microscopy observations indicated that cracks propagated parallel and perpendicular to the intermetallic layers in composites that had been treated 0.5 hour at 900°C (Fig. 3). Parallel cracks were observed on the border between TiCu layers and the zone consisted of $TiCu_4$ particles in Ti_3Cu_4+Ti_2Cu_3 matrix.

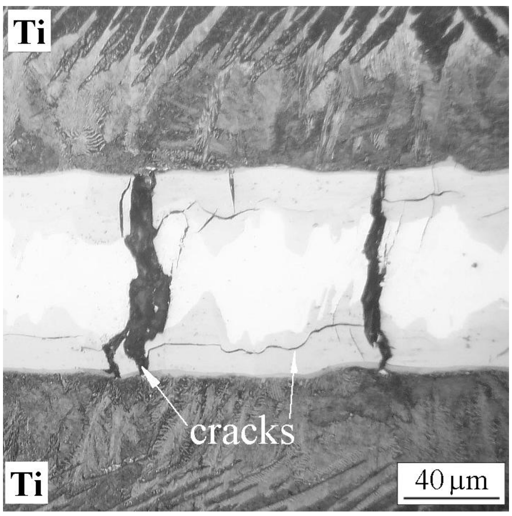

Fig. 3. Cracks in the specimen after 0.5 hour of treating at 900°C where testing was stopped prior to failure

Rys. 3. Pęknięcia w próbce uzyskanej po 30 minutach wygrzewania w 900°C powstałe w trakcie rozciągania

The presence of cracks in the intermetallic layers running mainly perpendicular to the metal layers was observed in composites after 5 hours of treating at 900°C (where titanide layers were composed primarily of the brittle TiCu phase). With permanent increase of the crack number in the layers of intermetallic phases the titanium layers gradually underwent the total external load. As a result the plastic flow that took place in the titanium layers was restricted to the regions between opposite cracks in the neighbouring layers of intermetallic phases. Thus a plastic strain of titanium was localised in shear bands. A damage of composites by shearing fracture of titanium layers was preceded by severe plastic deformation of these regions. This behaviour is typical of ductile-phase-toughened composites [4, 11].

As shown in Table 1, with an increase of the treating time at 900°C, the intermetallics grow, leading to an increase in volume fraction of the intermetallics. As a result, the yield strength of all investigated composites increases with increasing of the treating time. The tensile strength and the total strain at fracture decrease after long heat treatment (Table 2). On the one hand, it is a result of reduction of the volume fraction of titanium layers. On the other hand, it is attributable to a degradation of the intermetallic layers due to the presence of oxides. In fact, the vacuum in the experiments is not high enough (only 10^{-2} Pa) to prevent metal foils from oxidation at high temperature. Before a perfect interface is established between titanium and copper layers through interfacial reactions, oxygen diffuses along the interface to react with the metals to produce oxides. After 0.5 hour of treating at 900°C the oxides have no significant influence on the strength of the layers. EDS results show that after long heat treatment the oxides are captured in the growing intermetallic layers to form inclusions or voids. This results in the formation of weak points in the intermetallic layers, from which cracks would initiate easily, leading to premature failure of the layers during loading. It is hoped that by optimising the composite microstructure, the strengths of these composites may be improved considerably. It should be added that the composites exhibit a good cohesion between titanium layers and layers of intermetallic phases during tensile test.

CONCLUSIONS

The MIL composites were successfully formed by reaction synthesis of Ti and Cu foils. The microstructural characterisation indicated that TiCu is the predominant intermetallic phase. The formation of TiCu is dependent on treating time at 900°C and can be understood from the steps involved in another phases formation ($TiCu_4$, Ti_2Cu_3 and Ti_3Cu_4), which occurs through a series of the solid-liquid and the solid-state reactions. The tensile behaviour of the Ti-titanides laminated composites was examined. The results show that treating time at 900°C is a main factor determining properties because it leads to an increase in volume fraction of the intermetallics. Unfortunately, long heat treatment can also cause a degradation of the intermetallic layers by oxidation. Investigations indicate that the yield strength of all fabricated composites increases with increasing of the treating time. On the other hand, after 0.5 hours of treating the composites have higher ultimate tensile strength and higher strain at fracture. The results also show that the composites exhibit a good cohesion between titanium layers and layers of intermetallic phases during tensile test.

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