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Liquid-Phase Sintering of Al with Addition of Ti

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Abstract:

Samples made from aluminium-titanium elemental powder blend with titanium contents ranging from 5 to 20 at. % were prepared by die pressing and subsequent sintering under vacuum in a dilatometer quartz tube at 700, 800 and 900 ℃. Dilatometric and heating curves of sintering were analysed and discussed in connection with X-ray data and results from microstructural investigations.

Keywords: *Liquid phase sintering, Al with addition of Ti, Microstructure.*

Introduction

Further investigations of sintering of powder mixtures, due to its complexity and insufficient level of studies of the processes accompanying it, are required and it is one of the major areas for scientific research in powder metallurgy. Liquid-phase sintering of powder systems, in which intermetallic compounds are formed, deserves special attention.

In contrast to the traditional approach (selection of the quantitative ratio between high-melting and low-melting components in a mixture), we successfully evaluated liquid-phase sintering of mixtures on the basis of a component forming a liquid during sintering, namely aluminium [1-4]. The powders of transition metals Ti, Ni and Fe were used as alloying elements introduced into aluminium powder in quantities of 10-20 at. %.

In our research, with the help of a vacuum dilatometer, we studied the process of liquid-phase sintering of powder compacts made from Al-Ti elemental blends in which aluminium was the base of the compacts while titanium was the additive.

Experimental procedure

The content of the titanium powder in the mixtures varied from 5 to 20 at. %. Cylindrical compacts of powder mixtures with 10 mm in height and 10 mm in diameter were prepared with the initial porosity of 20%. For obtaining identical porosity and equal heights of green compacts, the mass of the sample from each mixture was previously calculated. Then,

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the samples were produced from the weighted portions by a method of two-sided pressing with a limiter of the top punch motion. For removal of adsorbed gases and moisture, prior to sintering, the compacts were exposed to one-hour annealing in vacuum at the temperature of 500 °C.

The quartz tube of the dilatometer containing the sample was placed in the furnace, which was preheated to the preset temperature [5]. This temperature in the dilatometer furnace was maintained constant throughout the complete sintering cycle. Due to alloy formation accompanied by a heat release inside compacts, the temperature of the samples temporarily rose above the furnace temperature at high sintering temperatures.For the study of the quantitative dependence of the final porosity of alloys on the concentration of titanium, the compacts were sintered in an ordinary vacuum furnace with a permanent pressure of 10^{-2} Pa at temperatures of 700, 800 and 865 °C for 1 hour. An investigation of the microstructure of alloys sintered in this furnace was made with a "Philips SEM 515" scanning electron microscope. Phase analysis was performed with a "Shimadzu XRD-6000" device.

Results

Dilatometry at 700 °C:. After reaching the melting point for aluminium (660 °C), the temperature of the compact containing 5 % Ti, while aluminium melts, remains constant during the heating process approximately for 7 minutes. Only afterwards, the temperature of the compact in the dilatometer tube reaches 700 °C, which is equal to the temperature in the tube itself (Fig. 1, curve 1').

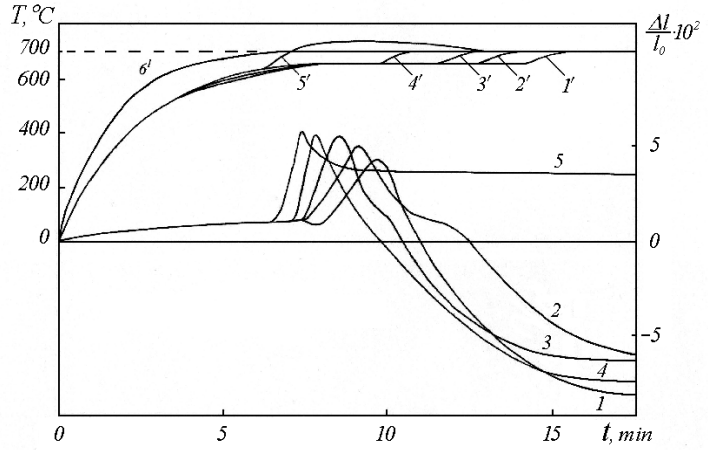

Fig. 1. Changes of relative linear dimensions (1-5) and temperature (1'-5') of Al-Ti compacts with the time of sintering at 700 °C: 1,1'– 5; 2,2'– 10; 3,3'– 12,5; 4,4'– 15; 5,5'– 20 at. % Ti; 6'– temperature in the dilatometer tube.

The initial stage of aluminium melting is accompanied by a slow growth of the sample size followed by volume shrinkage caused by formation of a liquid phase. Under the action of its own weight and the spring pushing the dilatometer measuring-rod, the sample finally loses its initial shape because of the excessive quantity of the liquid phase.

Based on the Ti-Al phase diagram, titanium and aluminium form two types of intermetallic compounds: TiAl and $TiAl_3$ [6]. However, the titanium content of 5% in the mixture is insufficient to form the solid phase intermetallic skeleton in the compact, which could prevent compact distortion. With a titanium content of 7.5 % and higher, the samples retain their initial shape, though a strain due to pressure transferred from the sample holder

affected the sample.

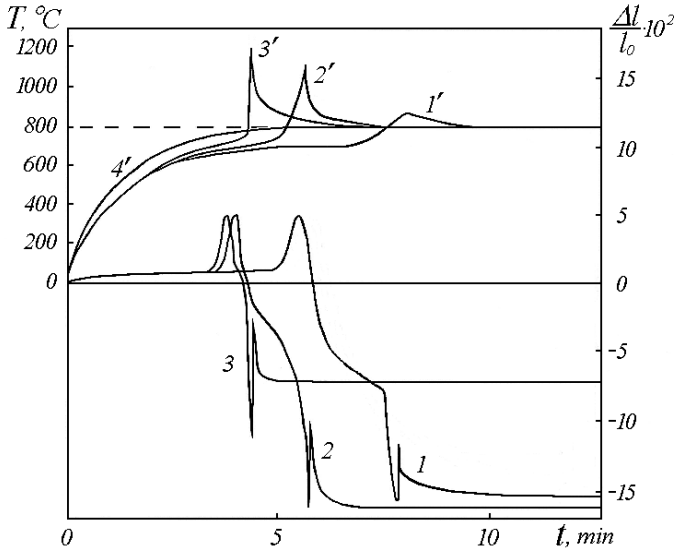

Fig. 2. Changes of relative linear dimensions (1-3) and temperature (1'-3') of Al-Ti compacts with the time of sintering at 800 °C: 1,1'– 12,5; 2,2'– 15; 3,3'– 20 at. % Ti; 4'– temperature in the dilatometer tube.

The special feature of sintering of compacts from the Al-20% Ti mixture at 700 °C in the dilatometer is that as a result of self-heating caused by alloy formation their temperature exceeds the temperature in the dilatometer tube. Besides, the compact growth exceeds its shrinkage in absolute magnitude (Fig. 1, curve 5).

Dilatometry at 800 °C: The temperatures in the dilatometer furnace and tube were kept constant at 800 °C. Dilatometric curves and temperature changes inside the compacts due to sintering effects are displayed in Fig. 2. Comparable to the sintering temperature of 700 °C, growing of compacts starts during melting of aluminium. However, this growth is replaced by shrinkage after some minutes. This effect occurs earlier with increasing the concentration of titanium in the sample. Owing to the appearance of a considerable quantity of the melt, the compact skeleton, mainly composed of Al_2O_3 oxide films and titanium particles, collapses. As a result of the phenomenon of solid phase particle rearrangement, sample shrinkage and distortion is observed till the sufficient quantity of the intermetallic compounds forms a new compact skeleton.

Dilatometry at 900 °C: Dilatometric curves of compacts, containing 12,5 and 20 at. % of titanium, sintered at the temperature of 900 °C are shown in Fig. 3. These curves are qualitatively comparable with the curves measured at 800 °C. However, the temperature peak is higher having the same content of titanium. For the Al-20 % Ti mixture the peak noticeably exceeds the temperature of 1200 °C.

Porosity of sintered alloys: The dependence of the final porosity of alloys sintered in the ordinary vacuum furnace at three temperatures as a function of the titanium concentration is shown in Fig. 4. During sintering, shrinkage was obtained for all compacts. The degree of shrinkage depended on the sintering temperature and titanium content. The higher the sintering temperature, the higher is the degree of compact densification. A maximum of the residual porosity of sintered alloys is observed for a titanium concentration of 7,5-10 at. %. With smaller titanium contents, the shrinkage increased. This fact indicates that a decrease of

titanium concentration reduces the content of intermetallic inclusions formed during sintering. However, if titanium contents in the mixture are greater than 10 %, shrinkage increases.

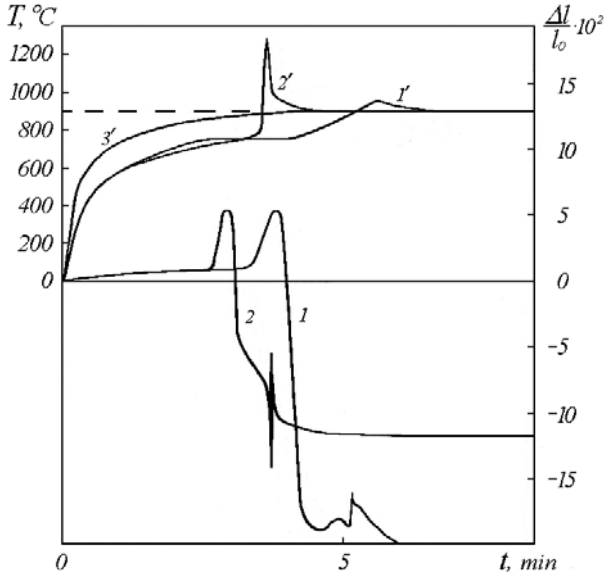

Fig 3. Changes of relative linear dimensions (1,2) and temperature (1',2') of Al-Ti compacts with the time of sintering at 900 °C: 1,1'– 12,5; 2,2'– 20 at. % Ti; 3'– temperature in the dilatometer tube.

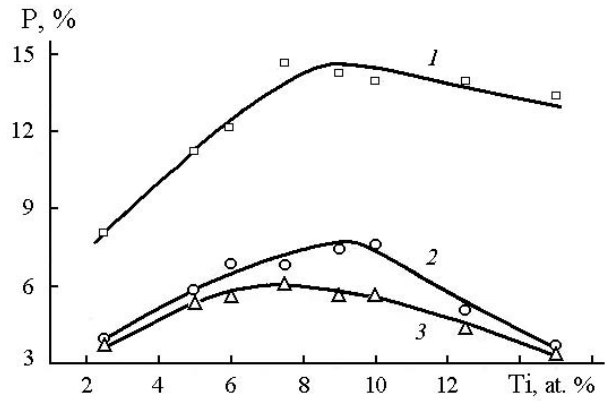

Fig. 4. Dependence of porosity of Al-Ti alloys sintered at 700 (1), 800 (2), and 865 °C (3) on the concentration of titanium.

This densification is presumably caused by the incipiency of the exothermic effect due to which the compact temperature reaches the order of 1200 °C and even higher. The exothermic effect is connected with the heat emission due to the alloy formation in the system with the negative enthalpy of mixing. The density of the sintered material increases with increasing sintering temperature.

X-ray phase analysis: The X-ray diffraction patterns of the alloys containing 10 and 20 at. % Ti and sintered under vacuum at 700 and 900 °C are illustrated in Fig. 5. The obtained results show that all alloys consisted of two phases: aluminium and the $TiAl_3$ intermetallic

compound. The intermediate TiAl phase is not detected. Judging by the intensity of reflections, there is a noticeably smaller quantity of aluminium in the alloy containing 20% Ti than in alloys with a smaller concentration of the additive, irrespective of sintering temperature. In the former case, the main part of aluminium has reacted with titanium forming the $TiAl_3$ compound. Titanium in the free form was not detected using the X-ray diffraction method also after sintering at 700 °C.

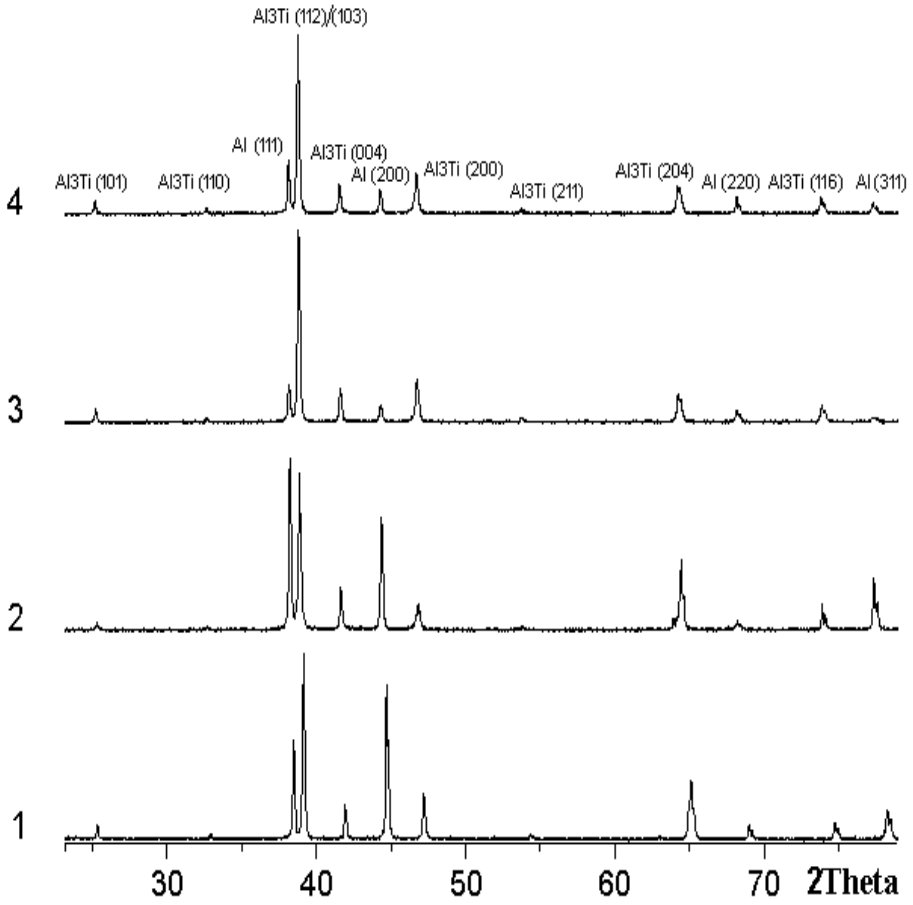

Fig. 5. X-ray diffraction patterns of the Al-Ti alloys containing 10 (1, 2) and 20 at. % Ti (3, 4) after sintering in the vacuum furnace at 700 (1, 3) and 900 °C (2, 4).

This may indicate that if titanium never remains after sintering as a pure element, its amount remaining in the alloys appears to be below the detection limit of the X-ray phase analysis.

Microstructure: The microstructure of samples with 10 at. % Ti after vacuum sintering at 700 and 900 °C is shown in Fig. 6. The two-phase constitution of alloys is not detected at small magnifications (×100). After sintering at 700 °C, the pores are irregular, and in most cases elongated in shape. As a result of sintering at the temperature of 900 °C the pores acquire a more spherical, equiaxial configuration. At higher magnifications (×1500) it is possible to see that the alloys consisted of two phases: the light phase represents the $TiAl_3$ intermetallic compound and the darker one is aluminium. One can also discern that pores are primarily concentrated in aluminium.

Discussion

With a titanium concentration in the mixture of more than 20%, the growth of compacts coincides in time with their intensive self-heating caused by the heat release during formation of intermetallic $TiAl_3$ [7]. As indicated by the dilatometric measurements (Fig. 1-3), a distinctive feature of sintering of Al-Ti mixtures containing less than 20 at. % of titanium is the temporary growth of compacts in volume. The reason for the swelling of compacts is the diffusion of aluminium atoms into titanium particles. Diffusion leads to an increased solid phase volume by 4 times due to formation of the $TiAl_3$ intermetallic. The quantity of the intermetallic compound formed in the compact and the amount of emitted heat increase with higher titanium concentration. As a result, the rate of sample growth and its absolute value slightly rise with an increase of the titanium content, and the beginning of the growth displaces towards smaller time intervals.

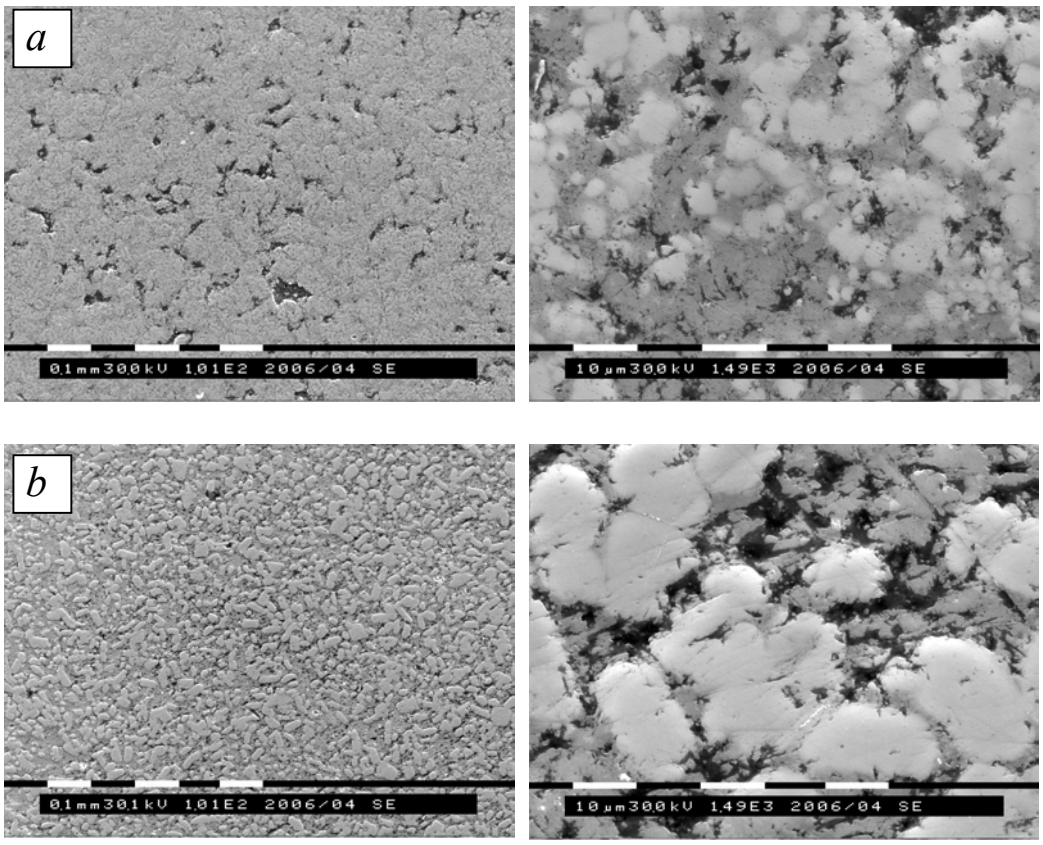

Fig. 6. Microstructure of alloys containing 10 at. % Ti after sintering at 700 (a) and 900 °C (b). Magnification: 100 (on the left) and 1500 (on the right).

As all the heat related to the reaction of the intermetallic compound formation is spent on the process of aluminium melting, no temperature splash is observed during sintering at 700 °C. The higher the content of titanium in the compact, the shorter is the period of melting (Fig. 1). In all cases, after formation of some critical amount of melt, a rearrangement of solid phase particles starts. As a result of the rearrangement, the compact growth is replaced by a fast shrinkage.

After the completion of aluminium melting, the emitted heat is available for heating of the compact especially for sufficient titanium concentrations. This heating, in turn, intensifies

the diffusion process and accelerates the formation of the intermetallic compound. In systems with a negative mixing enthalpy, the released heat in the diffusion zone raises the compact temperature and, thereby, even more increases the diffusion mobility of atoms accelerating their diffusion from the liquid phase into the solid. Thus, the process of the transition of atoms from the melt in the solid is a self-accelerating, or an avalanche-like, phenomenon in such systems. It is reasonable that the compact growth and the temperature peak during this exothermic effect coincide in time with each other. This is the case in the course of densification process during sintering at 800 and 900°C (Fig. 2 and 3). As a result, shrinkage is interrupted by a new compact growth, which took place during a short time. This growth becomes apparent on the dilatometric curves as an acute peak, to which a short-term, but noticeable on value, shrinkage precedes.Then, after the fast growth, the shrinkage continues with an ordinary rate.

The titanium exhaustion causes a gradual drop of the sample temperature to the level of the temperature in the dilatometer tube. In this case, the rearrangement of solid phase particles is finished with formation of a new compact skeleton. The new skeleton, besides oxide films, consists of intermetallic inclusions, the average size does not actually depend on the concentration of titanium at the same sintering temperature and is determined completely by the temperature of sintering. The higher is the sintering temperature, the larger are the intermetallic inclusions. The average size of $TiAl_3$ particles in the alloys sintered at 700 °C is 3 mμ, at 800 °C it is 7 mμ, and at 900 °C it is 15 μm. Obviously, with increasing of sintering temperature, enlargement of the $TiAl_3$ particles occurs because of the solution-reprecipitation phenomenon.

Conclusions

1. The compacts based on aluminium powder containing several amounts of titanium powder (from 5 to 20 at. %) are capable of exothermal reaction sintering at temperatures above the melting point of aluminium.
2. Higher titanium contents in the mixture lead to a higher temperature of the sample during sintering due to a reaction of aluminium from the melt with titanium particles.
3. Exothermic sintering is accompanied by swelling at the aluminium melting point with the consequence of substantial shrinkage brought about by the process of rearrangement of solid phase particles in the melt.
4. Compact growth is connected with diffusion of aluminium atoms from the melt into particles of titanium forming the $TiAl_3$ compound. The short-term renewal of the compact growth in the stage of shrinkage is the result of intensive intermetallic formation during sintering at high temperatures.
5. The TiAl intermetallic compound is not formed during sintering of the Al-Ti system.

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Садржај: *Узорци од мешавине прахова алуминијум-титана при чему је садржај титана између 5 и 20% су припремљени пресовањем па синтеровањем у вакуму у дилатометру са кварцом цеви на 700, 800 и 900°C. Дилатометријске криве синтеровања и криве загревања током синтеровања су анализиране и дискутоване у вези са рентгенским подацима и резултатима микроструктурних анализа.*
Кључне речи: *Синтеровање у присуству течне фазе, Al са додатком Ti, микроструктура.*