X-RAY DIFFRACTION OF A Ti-Ta-Nb ALLOY PROCESSED BY SEVERE PLASTIC DEFORMATION

Andreea Daniela VULCAN[1], Doina RĂDUCANU[2], Vasile Dănuț COJOCARU[3]
Ion CINCA[4], Lucia ANGELESCU[5]

În prezenta lucrare au fost investigate caracteristicile structurale ale aliajului Ti-25Ta-25Nb supus deformării plastice severe (SPD) prin procedeul de laminare acumulativă (ARB) prin tehnica de difracție cu raze X (XRD). Investigațiile XRD au dezvăluit faptul că faza inițială, β, se divide în două sub-faze β (β1 si β2) printr-un mecanism de tensiune indus, ca urmare a intensității de deformare mari.

Stabilitatea fazei β a fost dictată de compoziția aliajului şi influențează comportamentul deformări. Un aspect general este faptul că, gradul de deformare al straturilor nu este omogen. Prin urmare toate probele prezintă o textură puternică, fapt ce va fi prezentat în următorul articol, indicând faptul că aliajele de Ti- β - în acest caz, aliajul Ti-25Ta-25Nb - pot fi folosite în noile aplicații medicale, datorită proprietăților lor favorabile.

In the present paper the structural characteristics of Ti-25Ta-25Nb alloy subjected to severe plastic deformation (SPD) by accumulative roll-bonding (ARB) process have been studied by XRD investigations. The XRD investigations revealed that the initial β phase is divided in two β sub-phases (β1 and β2) through a stress induced mechanism, as a result of the high intensity deformation.

The β phase stability was dictated by the alloy composition and influenced the deformation behaviour. A general aspect is that the deformation degree of the layers is not homogeneous. Therefore, all samples present a strong texture, which will be presented in a future article, indicating that the promising β-Ti alloys – in this case Ti-25Ta-25Nb alloy – can be used in new medical applications, due to their favourable properties.

Keywords: titanium alloy, beta phase stability, X-ray diffraction, pole figures

---

[1] PhD Student, Materials Science and Engineering Faculty, University POLITEHNICA of Bucharest, Romania, e-mail: Vulcan_andreea@yahoo.com

[2] Professor, Materials Science and Engineering Faculty, University POLITEHNICA of Bucharest, Romania, e-mail: doina.raducanu@mdef.pub.ro

[3] Lecturer, Materials Science and Engineering Faculty, University POLITEHNICA of Bucharest, Romania, e-mail: dan.cojocaru@mdef.pub.ro

[4] Reader, Materials Science and Engineering Faculty, University POLITEHNICA of Bucharest, Romania, e-mail: ion.cinca@mdef.pub.ro

[5] Reader, Materials Science and Engineering Faculty, University POLITEHNICA of Bucharest, Romania, e-mail: lucia.angelescu@mdef.pub.ro

1. Introduction

Titanium and its alloys are used for applications as biomaterials in biomedical area, due to a favourable combination of mechanical, chemical and physical properties, such as low density, high mechanical resistance, excellent corrosion resistance and good biocompatibility. A good choice was to add niobium (Nb) and tantalum (Ta) to titanium (Ti), since both acts as β-stabilizers, which improve the mechanical properties [1].

There is considerable research focussed on the development of new β titanium alloys for biomedical applications due to their favourable properties, combined with their chemical compositions, where Ni and other toxic elements are absent.

The SPD procedures may be applied to a great variety of materials with advanced purity and high mechanical properties [2]. Being a SPD method, ARB involves a severe deformation of metal sheets, without modification of overall material thickness. ARB has been previously applied to grain refinement of commercial purity Ti; however, there is very little information on the ARB of metastable β Ti alloys in the literature. Benefits of ARB include the use of conventional roll forming equipment and the ability to produce bulk UFG sheets with dimensions limited only by the capacity of the rolls. ARB involves stacking of two metal sheets and rolling them together, in order to refine the grain structure and make them to adhere through the roll-bonding process. The rolled sheet is then half sectioned, cleaned, stacked and afterwards the process is repeated. Thus, a very large plastic strain can be achieved over a number of cycles [3]. An important aspect of ARB procedure is the layer bonding process. The adhesion strongly depends not only on the surface treatment and rolling temperature, but also on the initial geometry of the sample.

The deformation mechanisms in beta-titanium alloys can widely vary, depending on the specific alloy composition and beta-grain size. These mechanisms include dislocation glide and climb, twinning and stress induced shear-type phase transformations. In turn, the specific deformation mechanism often has a strong impact on workability. Solid-solution beta-titanium alloys may exhibit good cold workability, thus making easier the part fabrication. However, when twinning and shear-type phase transformations predominate, cold workability can be markedly reduced.

Among these processes, the ARB process developed by Saito et al. (1999) has several advantages over other SPD processes: (1) no need for forming facilities with large load capacity and expensive dies; (2) high productivity; (3) unlimited production. Due to its capability as a continuous process, only ARB is appropriate for manufacturing nanocrystalline and ultrafine grained sheets and plates, which are the most widely used forms of commercial products [4].

In the present work the Ti–25Nb–25Ta alloy was analyzed, in order to validate its application as a biomaterial for biomedical industry. Physical and chemical characteristics of this alloy were evaluated through X-ray diffraction and scanning electron microscopy. The alloy was compressed up to ~ 87% reduction in height.

2. Methods

2.1. Alloy synthesis

The investigated alloy has been produced using a vacuum induction levitation melting furnace type FIVES CELLES, with a nominal power of 25 kW and a melting capacity of 30 cm^3, starting from elemental components. The resulted chemical composition in wt.% was: 50%Ti; 25%Ta; 25%Nb.

Firstly, the alloy was thermo-mechanically processed by cold-rolling and recrystallization, as seen in figure 1. One can observe that a first cold-rolling was performed up to a deformation degree of 82 %, using a Mario di Maio LQR 120AS rolling-mill, at a rolling speed of 2,4 m/min. The initial sample had 2.11 mm thickness. After the first cold-rolling, a recrystallization heat treatment was performed. The recrystallization treatment was carried out in a GERO SR 100X500/12 furnace. Recrystallization parameters were as follows - temperature: 850^0C; duration: 0.15 minutes; treatment media: vacuum; cooling media: air. A second cold-rolling was then performed, with a deformation degree of 67%, in order to obtain the precursor samples for ARB processing.

The accumulative roll-bonding process consisted in 3 cycles and the ARB process was conducted at ambient temperature, using the same Mario di Maio LQR 120AS rolling-mill, at a rolling speed of 2,4 m/min.

2.2. SPD process

Accumulative roll bonding (ARB) is one of the severe plastic deformation (SPD) processes, which can produce bulk ultrafine grained (UFG) metallic materials. The process is represented systematically in figure 2. Stacking of sheets and conventional roll-bonding are repeated during the process. Firstly, a strip is neatly placed on the top of another strip. The interfaces of the two strips are surface-treated in advance, in order to enhance the bonding strength. The two layers are joined together by rolling, as in the conventional roll-bonding process. Then, the rolled material is cut lengthwise into two halves. The sectioned strips are again surface treated, stacked and roll-bonded. In principle these procedures can be limitlessly repeated, so that very large plastic strain can be applied to the material.

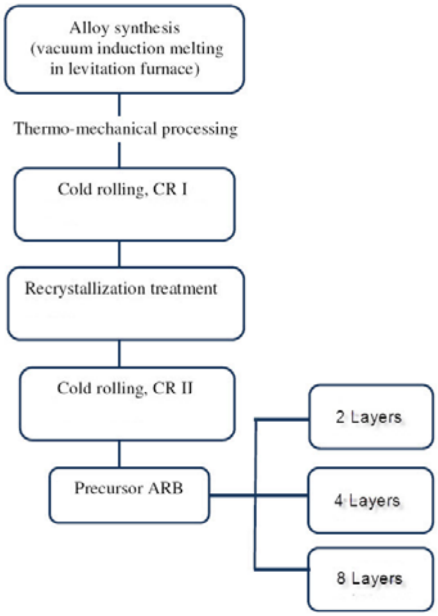

Fig. 1. Schematic representation of the experimental program

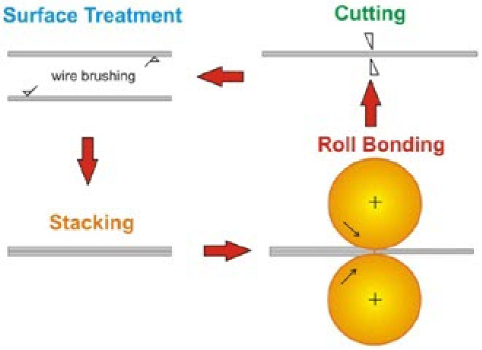

Fig. 2. Schematic illustration showing the principle of the ARB process

2.3. X-ray diffraction

The phase constitution of Ti-25Ta-25Nb alloy was determined using an X-ray diffractometer type Philips PW 3710, in Bragg-Brentano θ-2θ geometry.

The X-ray diffraction patterns were recorded on the RD and TD plane surfaces of the ARB-processed sheets. The XRD characterization was performed using a classical diffractometer Philip PW 3710, type Bragg-Brentano θ-2θ, with negligible instrumental broadening using Cu Kα radiation ($\lambda = 0.15406$ nm) in the range $2\theta = 30^0$–80^0, using a step size of 0.02, 40kV and 30mA.

3. Results and Discussion

3.1. ARB processing

Severe plastic deformation (SPD), which is a method that consists of applying very large strains, often to a bulk sample, proved to be a very effective way of fabricating UFG materials. UFG bulk metals and composites are considered very attractive structural materials, since they are significantly stronger than their coarse - grained counterparts and due to their good ductility. Additionally, UFG metals have improved corrosion resistance and potential for superplasticity at high strain rates and low temperatures. As a result of increasing the number of ARB cycles, the thickness of the layers decreases and the number of layers per unit thickness increases. This layer refinement affects the strength of the produced multilayer material, via the so-called Hall–Petch relationship.

The starting precursor sheets were firstly deformed to approx. 50% reduction in thickness, by conventional rolling. The resulted stack was cut in half - length, surface treated and stacked again, resulting the first ARB cycle. The second and third ARB cycles were performed in the same manner. The resulted deformation degree after each ARB cycle was approximately 43%. Table 1 shows the number of layers and the layer thickness after each cycle.

Table 1

Roll Cycle data for the ARB- processed Ti-25Ta-25Nb alloy.

Cycles	Layers	Layer thickness (mm)	
		Initial	Final
Precursor	1	0.09	-
1 (ARB)	2	0.09	0.07
2 (ARB)	4	0.07	0.05
3 (ARB)	8	0.05	0.02

3.2. XRD analysis

Specimens for XRD were prepared by polishing. XRD was carried out using an X-ray diffractometer with Cu-Kα radiation, at a scanning speed of 0.03 m/s. The jig fixed with the specimens was placed into the chamber of the X-ray diffractometer and then subjected to X-ray diffraction analysis (XRD). In XRD the accelerating voltage, the current intensity and the scanning angle (2θ) were as follows: 40 kV, 30 mA and 30°–80°.

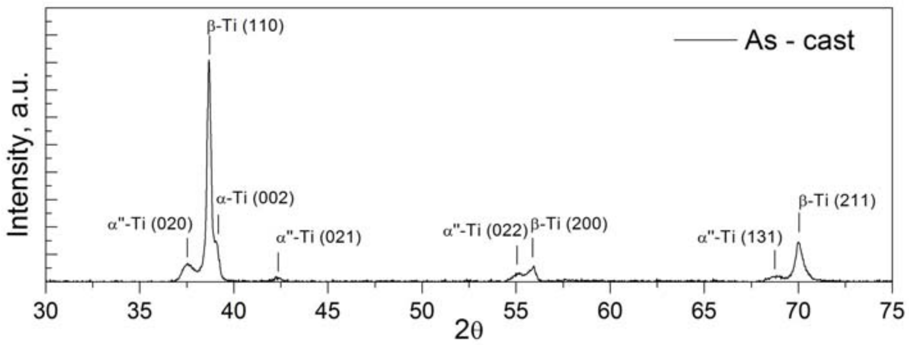

Fig.3. X-ray diffraction pattern of as-cast Ti-25Ta-25Nb alloy

The XRD profile for the as - cast sample (figure 3) shows the presence of martensite phase (α”) and the presence of α and β phases. Diffraction was carried out in order to observe the main peaks of the β phase (1 1 0), (2 0 0) and (2 1 1).

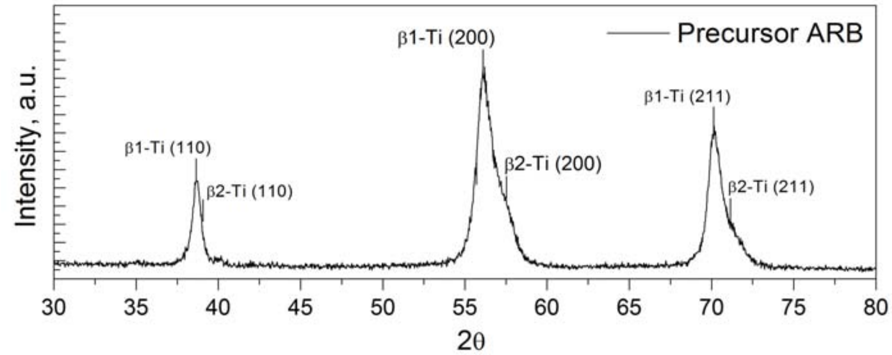

Fig.4. X-ray diffraction pattern of precursor samples for ARB processing

The XRD patterns shown in figure 4 is representative for precursor samples for ARB processing; one can observe that the α” martensite and α phase disappear and only the β phase peaks can be observed. Another observation is that the initial β phase is divided in two β sub-phases (β_1 and β_2). The relative peak heights, in particular the large (220) and (211) peaks, suggest that additional β sub-phase was formed through a stress induced mechanism, as a result of the intense deformation.

In Fig. 5 (2 layers) one can observe that the most important phase is β_1, while in figure 6(4 layers) phases β_1 and β_2 are almost equal. For 8 layers (figure 7) the situation is changed, because the β_2 sub-phase becomes the most important, having the highest peaks. The generation of defects during high intensity deformation, such as grain boundaries, dislocations and vacancies may enhance the splitting of initial β phase in two β sub-phases (β_1 and β_2). The XRD pattern shows significant peak broadening, indicative of fine grain size in nanometric range.

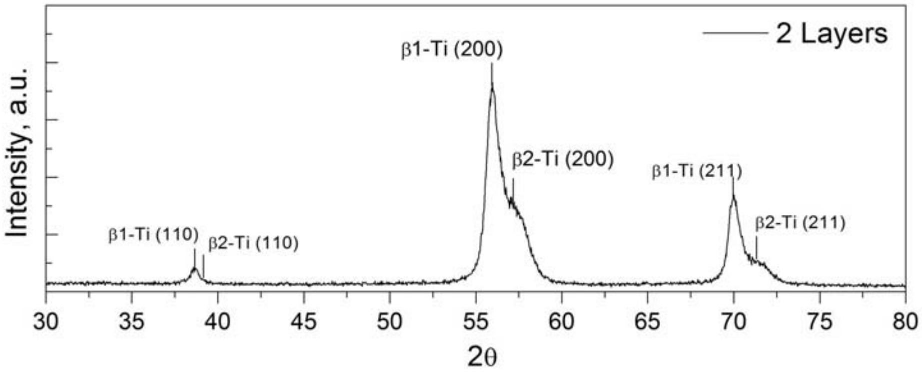

Fig.5. X-ray diffraction pattern for 2 layers ARB processed alloy

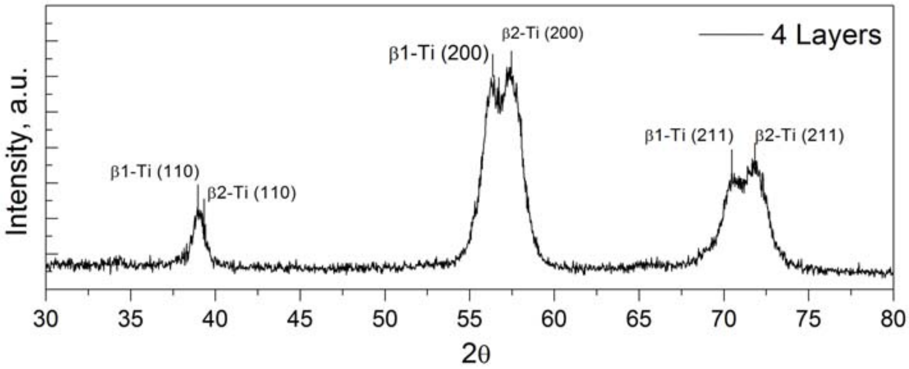

Fig.6. X-ray diffraction pattern for 4 layers ARB processed alloy

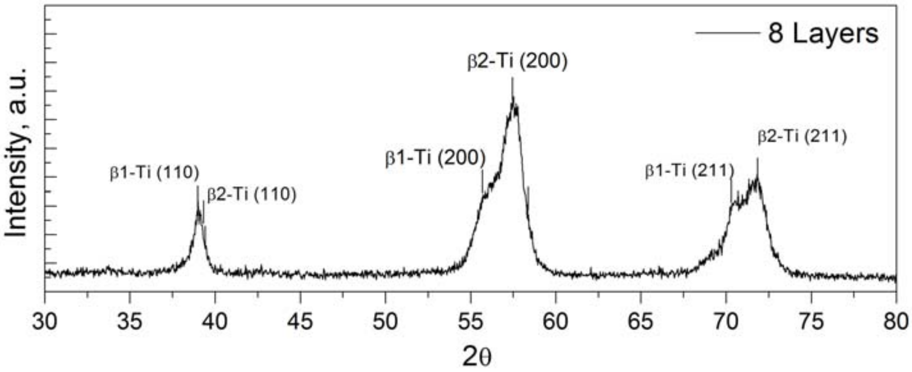

Fig.7. X-ray diffraction pattern for 8 layers ARB processed alloy

A "splitting" of the β - phase reflections was observed for the first time on XRD curve for all the samples, which shows that a separation reaction has taken place, leading to the formation of two b.c.c. β-sub phases. Some researchers have reported that the decomposition of the metastable β-phase in titanium alloys proceeds through a phase separation reaction, leading to the formation of solute rich (β_1) and solute lean (β_2) b.c.c. phases and the evidence for such a reaction was broad and diffuse for the β-phase reflections. In general, no peak splitting can be achieved using X-ray diffraction technique, since the difference in the lattice parameters of β_1 and β_2 is relatively small (a= 3.302 A for β_1 and a=3.262 A for β_2).

4. Conclusions

The XRD investigations revealed that the initial β phase is divided in two β sub-phases (β_1 and β_2) through a stress induced mechanism, as a result of the high intensity deformation. Generation of defects during high intensity deformation, such as grain boundaries, dislocations and vacancies, may enhance the splitting of initial β phase in two β sub-phases (β_1 and β_2).

The β phase stability was dictated by the alloy composition and influenced the deformation behaviour. A general aspect is that the deformation degree of the layers is not homogeneous. Therefore, all samples present a strong texture, which will be presented in a future article, indicating that the promising β-Ti alloys – in this case Ti-25Ta-25Nb alloy – can be used in new medical applications, due to their favourable properties.

REFERENCES

[1] *L. R. Ribeiro, R. C. Junior, F. F. Cardoso, R. B. F. Filho, L. G. Vaz*, Mechanical, physical, and chemical characterization of Ti–35Nb–5Zr and Ti–35Nb–10Zr casting alloys, Journal Materials Science, **20**, 2009, 1629-1636, Sa˜o Paulo State University, Brazil;

[2] *D. Răducanu, V. D. Cojocaru, I. Cinca, I. Ichim, A. Sichin*, Materials development on the nanoscale by Accumulative Roll Bonding procedure, Journal of optoelectronics and advanced materials, **9**, 2007, 3346 – 3349, University Politehnica of Bucharest, Romania;

[3] *D. Kent, G. Wang, Z. Yu. X. Ma, M. Dargusch*, Strength enhancement of a biomedical titanium alloy through a modified accumulative roll bonding technique, Journal of the mechanical behaviour of biomedical materials, **4**, 2011, 405 – 416, University of Queensland, Brisbane QLD 4072, Australia, Biomaterial Research Centre, Northwest Institute for Nonferrous Metal Research, Xian 710016, China;

[4] *Y. Saito, N. Tsuji, H. Utsunomiya, T. Saka, R. G. Hong*, Ultra-fine Grained Bulk Aluminum Produced by Accumulative Roll-bonding (ARB) Process Scripta Materialia, **39**, 1998, 1221–1227, Departament of Materials Science and Engineering, Osaka University, Japan;

[5] *Y. Saito, H. Utsunomiya, N. Tsuji, T. Sakai*, Novel Ultra-high Straining Process for Bulk Materials-development of the Accumulative Roll-bonding (ARB) Process, Acta Materialia, **47**, 1999, 579–583, Division of Materials Science and Engineering, Graduate School of Engineering, Osaka University, Japan.