X-RAY DIFFRACTION INVESTIGATIONS OF A Ti-29Nb-9Ta-10Zr THERMO-MECHANICAL PROCESSED ALLOY

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In the present paper, a Ti-29Nb-9Ta-10Zr (wt.%) alloy was thermo-mechanical processed and analysed in order to investigate structural changes occurred during processing. The X-ray diffraction investigation was used in order to characterize obtained structures during thermo-mechanical processing, using a Philip PW 3710 diffractometer. Data concerning alloys component phases, average coherent crystallite size and micro-strain at crystalline lattice level was obtained.

Keywords: Titanium alloy, Thermo-mechanical processing, X-ray diffraction

1. Introduction

Titanium and its alloys have been widely used in medicine since the 1960s because of their known biocompatibility, superior mechanical properties, low density and remarkable chemical stability.

During the last decades, many researches were focused on titanium alloys used in biomedical applications, due to their favourable combination of mechanical resistance with physical and chemical properties such as: low density, high mechanical resistance, excellent corrosion and good biocompability. The addition of non-toxic elements such as niobium (Nb), tantalum (Ta) and zirconium (Zr) acts on β-phase stability and also is a way to improve mechanical properties [1-3].

Complementary, thermo-mechanical processing is often applied on titanium alloys in order to obtain a desired combination of mechanical properties based on various complex structures.

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In the case of β-Ti alloys used in medical applications, especially in osseous implantology, a low elastic modulus is desired. In order to decrease the elastic modulus, microstructures consisting in a mixture of both β-Ti and α″-Ti phases must be obtained, due to the fact that α″-Ti phase exhibits a lower elastic modulus in comparison with parent β-Ti phase [4].

In this paper a Ti-29Nb-9Ta-10Zr (wt.%) β-Ti alloy was thermo-mechanical processed in different structural states, by homogenization, heavy cold-rolling, controlled recovery by recrystallization and annealing and investigated by X-ray diffraction, in order to determine data concerning alloys component phases, average coherent crystallite size and internal average micro-strain.

2. Methods

The investigated alloy has been produced using a vacuum arc induction melting in levitation furnace FIVES CELES with nominal power 25kW and melting capacity 30 cm^2, starting from high purity elemental components. Resulted chemical composition, in wt. %, was: 52%Ti; 29 Nb; 9Ta; 10Zr.

The thermo-mechanical processing route applied on Ti-29Nb-9Ta-10Zr alloy is presented in figure 1. From as-cast alloy samples were cutted in order to process them by cold-rolling. Prior to first cold-rolling the samples were subjected to a homogenization heat treatment in order to obtain an internal structure without major casting defects, such as segregation volumes. Heat treatment parameters were: homogenisation temperature: 950^0 C; treatment duration: 360 min, heating medium: high vacuum; cooling medium: furnace.

After homogenization treatment a cold-deformation process was applied in order to refine internal microstructure. Cold-rolling deformation was used as deformation procedure, using a Mario di Mario LQR120AS laboratory roll-milling machine, with a rolling speed of 2.4 m/min. Total thickness reduction applied during cold-rolling was about 60%.After cold-rolling a recrystallization treatment was applied. For the recrystallization heat treatment a GERO SR 100X500/12 – high temperature furnace has been used. The parameters of the recrystallization were as follows: recrystallization temperature: 780^0 C; recrystallization duration: 0.5 min, treatment media: argon; cooling media: water.

The thermo-mechanical processing was continued with a second cold-rolling deformation was applied with a 90% total thickness reduction.After the second cold-rolling, a recrystallization treatment with water quenching was applied, in order to remove the effects of strain-hardening and to obtain a structure consisting of β-Ti and α″-Ti phases. The second recrystallization treatment has been performed in the same conditions as the first recrystallization treatment.

A final aging heat treatment was applied with the aim to reduce internal micro-strain of both β-Ti phase and α″-Ti phases and also to consolidate both phases.

Aging treatment parameters were: aging temperature: 400^0 C; duration: 90 min, treatment medium: high vacuum; cooling medium: furnace.

All heat treatments were performed using a GERO SR 100X500/12 – high temperature furnace.

The XRD characterization was performed on all structural states, using a Philip PW 3710 diffractometer, in Bragg-Brentano θ-2θ geometry, using Cu Kα radiation (λ = 0.15406 nm). All recorded XRD spectra were simulated and fitted. The XRD spectra simulation was performed using MAUD v2.33 software package, in order to calculate phases crystalline lattice parameters. The fitting procedure was performed using PeakFit v4.12 software package, in order to determine for each peak the position, intensity and peak broadening - FWHM (*Full Width at Half Maximum*). A pseudo-Voigt diffraction line profile was used in fitting procedure.

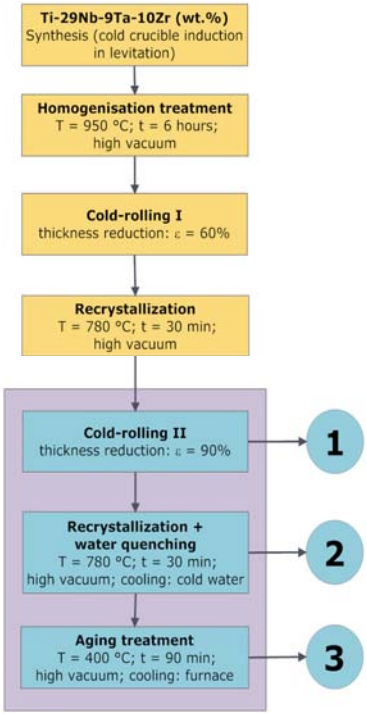

Fig. 1. Schematic representation ofthermo-mechanical processing routeapplied to Ti-29Nb-9Ta-10Zr alloy.

3. Results and discussion

3.1.Component phases

The XRD characterization of the Ti-29Nb-9Ta-10Zr alloy was performed in the range 2θ = 30° -80°. Data concerning alloys component phases, average coherent crystallite size and internal average micro-strain was obtained. In the case of β titanium alloys the parent β-Ti phase can be transformed in hexagonal martensite (α′) or orthorhombic martensite (α″). Orthorhombic α″ martensite can be formed by transformation of the parent β phase during recrystallization or by stress-induced transformation [5]. The presence of α"-phase in the β-Ti alloys has a good influence on the mechanical properties, especially on elastic modulus, decreasing it's value.

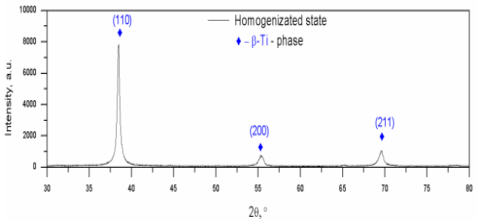

Fig.2. XRD spectra of homogenized Ti-29Nb-9Ta-10Zr alloy.

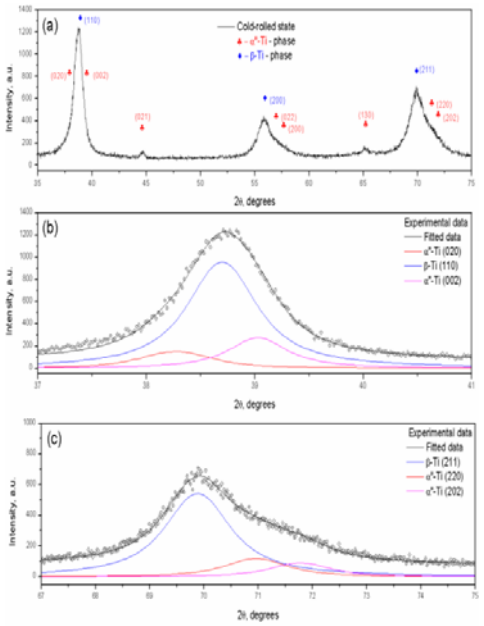

Fig. 3. XRD spectra of 90% cold-rolled Ti-29Nb-9Ta-10Zr alloy (a),detailed zoom of (110) β -Ti peak (b) and detailed zoom of (211) β -Ti peak (c).

The homogenised, 90% cold-rolled, recrystallized and aged structural states of the studied alloy were investigated using XRD diffraction. Recorded XRD spectra are presented in figure 2 - 5. As observed in figure 2, in the case of homogenized state the microstructure consists of a single β-Ti phase. The β-Ti phase was indexed in body centered cubic system – *Im-3m*. Observed β-Ti diffraction peaks

were as follows: (110), (200) and (211). In the case of 90% cold-rolled state (figure 3) the microstructure consists of a mixture of β-Ti and α″-Ti phases, the α″-Ti phase being induced by stress-induced transformation. The α″-Ti phase was indexed in orthorhombic system - *Cmcm*.Major diffraction peaks observed in the case of α″-Ti phase were as follows: (020), (002), (022), (002), (220), and (202).

In the case of recrystallized (figure 4) and aged (figure 5) states same β-Ti and α″-Ti peaks are observed, with the differences in peaks intensities and broadening, suggesting consolidation of both phases as a result of recrystallization and aging (increase of grain-size and decrease of average internal micro-strain).

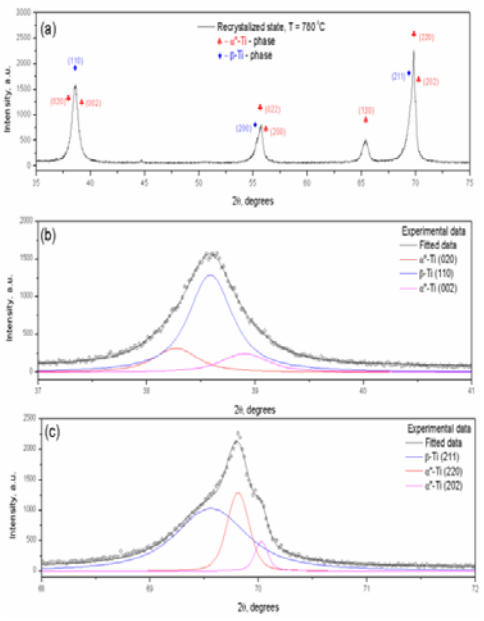

Fig. 4. XRD spectra of recrystallized Ti-29Nb-9Ta-10Zr alloy (a),detailed zoom of (110) β-Ti peak (b) and detailed zoom of (211) β-Ti peak (c). zoom of (211) β-Ti peak (c).

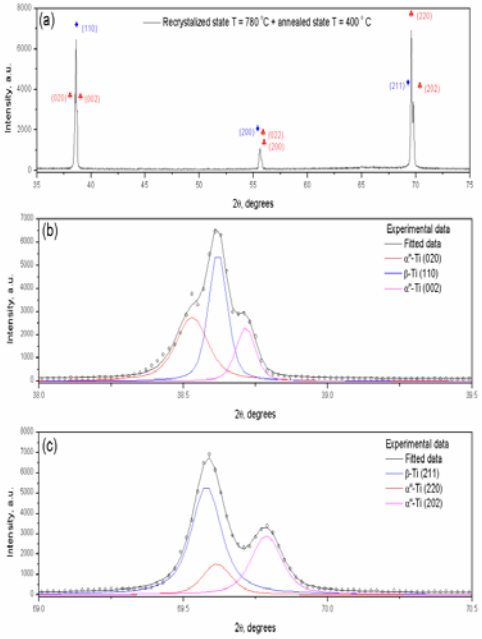

Fig. 5. XRD spectra of aged Ti-29Nb-9Ta-10Zr alloy (a),detailed zoom of (110) β-Ti peak (b) and detailed zoom of (211) β-Ti peak (c).

3.2.Lattice parameters

All XRD spectra's were fitted in order to deconvolute observed cumulative diffraction peaks and to obtain for each constitutive peak it's position, intensity and broadening.

In the case of 90% cold-rolled state (structural state 1), as observed in figure 3 (b and c), in 2θ = (30 - 40)° scattering domain the following peaks sequence is observed: α″(020) – β(110) – α″(002). In the case of 2θ = (60 - 70)° scattering domain the observed peaks sequence is as follow: α″(220) – β(211) –

α″(202).Same peaks sequences are observed also in the case of recrystallized state and annealed state, as can be seen in figure 4 (b and c) and figure 5 (b and c).

For each diffraction peak it was obtained it's position, intensity and broadening.Using obtained data concerning peaks position, computations were made in order to calculate crystalline lattice parameters in the case of both β-Ti and α″-Ti phases. Obtained data were as it follows:

- Homogenized state:
  - β-Ti phase: a = 3,31 Å;
- 90% cold-rolled state :
  - β-Ti phase: a = 3,29 Å;
  - α″-Ti phase: a = 3,21 Å; b = 4,70Å; c = 4,59Å;
- Recrystallized state :
  - β-Ti phase: a = 3,30Å;
  - α″-Ti phase: a = 3,28Å; b = 4,68Å; c = 4,65Å;
- Aged state:
  - β-Ti phase: a = 3.29Å;
  - α″-Ti phase: a = 3.28Å; b = 4.67Å; c = 4.65Å;

Analysing the α″-Ti phase lattice parameters, can be observed that in the case of 90% cold-rolled state the α″-Ti phase was obtained due to the *stress-induced transformation*, while in the case of recrystallized and annealed states was obtained due to the *temperature-induced transformation*.

3.3.Crystallite-size and micro-strain

Using obtained data concerning peaks broadening (FWHM - *Full Width at Half Maximum parameter*), further computations were made in order to calculate coherent grain-size domains and internal average micro-strain in the case of both β-Ti and α″-Ti phases, for all investigated structural states.

In order to calculate coherent crystallite size and internal average micro-strain the Williamson-Hall equation [6] was used:

$$FWHM \cdot cos\theta = 0.9 \cdot \frac{\lambda}{D} + 4 \cdot \varepsilon \quad (1)$$

where: FWHM parameter is the broadening of the diffraction peak measured as *Full Width at Half Maximum,* θ is the Bragg angle, λ is the wavelength of the X-ray radiation, ε is the average micro-strain and *D* the average dimension of crystallites (coherent grain-size).

Calculated grain-size and internal average micro-strain values are presented in table 1.

It can be observed that in the case of homogenized state the β-Ti phase show a grain-size close to 77.2 nm while the internal average micro-strain reaches a value close to 0.11%. A sharp decrease in coherent grain-size of β-Ti phase is noticed, in the case of cold-rolled state, to a value close to 37.48 nm, while the internal average micro-strain increase to a value close to 0.86%. Newly formed α″-Ti phase show acoherent grain-size close to 60.29 nm while its internal average micro-strain reaches 0.71%, in order to accommodate β→α″ stress-induced transformation.

The result of recrystallization and annealing treatments consists in small increments of coherent crystallite-size for both, β-Ti and α″-Ti phases, while the internal average micro-strain decreases dramatically.

As expected, maximum average micro-strain reaches a maximum, in the case of 90% cold-rolled state, close to 0.86%. In the case of - α″-Ti phase the maximum of average micro-strain is obtained in the case of stress-induced transformation, reaching 0.71%. The α″-Ti phase crystallite size is higher in the case of 90% cold-rolled states – close to 61, in comparison with annealed state - close to 45 nm and recrystallized state – close to 11 nm.

Table 1

Calculated crystallite size (grain-size)and internal average micro-strain

Structural state	β-Ti - phase		α″-Ti - phase	
	ε [%]	D [nm]	ε [%]	D [nm]
Homogenized state	0.11	77.2	-	-
90% Cold-rolled state	0.86	37.48	0.71	60.29
Recrystallized state	0.37	106.66	0.45	10.58
Annealed state	0.04	173.32	0.06	44.73

4. Conclusions

The present study investigates the microstructures and the mechanical properties of a Ti-29Nb-9Ta-10Zr (wt.%) alloys in order to investigate structural changes occurred during recrystallization and annealing treatment of 90% cold rolled. The conclusions of this study are summarized as follows.

The XRD investigations applied on Ti-29Nb-9Ta-10Zr (wt.%) alloy, thermo-mechanical processed, revealed that the phase components for all the studied

samples, excepting homogenized state, consist of a mixture of β-Ti phase and α″-Ti phase.

The α″-Ti phase is obtained by stress-induced transformation in the case of 90% cold-rolled state while in the case of recrystallized state is obtained by temperature-induced transformation.

By applying an intense cold-rolling deformation processing a nanometrical microstructure, consisting in a mixture of β-Ti and α″-Ti phases, is obtained; this nanometrical microstructure is retained also after recrystallization and aging treatments.

Acknowledgement

The authors wish toacknowledge the support of UPB Project no. POSDRU/107/1.5/S/76903 - "*Training of future researchers experts by Ph.D. scholarships*" and "*Functionalized super-elastic alloys for load-bearing implants*" project code ISA MANUNET-2012-1003 from UEFISCDI - Romania.

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