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Processing of Commercial Purity Titanium by ECAP Using a 90 Degrees Die at 350 °C Temperature

Využití technologie ECAP pro zpracování titanu při teplotě 350 °C v matrici s úhlem 90°

doc. Ing. Miroslav Greger, CSc.; **Ing. Rostislav Kawulok, Ph.D.**

VŠB – Technical University of Ostrava, Faculty of Metallurgy and Materials Engineering., Regional Materials Science and Technology Centre, 17. listopadu 15/2172, 708 33 Ostrava-Poruba, Czech Republic

Experiments were conducted to determine, whether it is feasible to process commercial purity (CP) titanium (Grade 2) by equal-channel angular pressing (ECAP) at 350 °C temperature with a die having an internal angle of 90°. Billets were successfully processed by up to 8 passes using a pressing speed of 3.0 mm·s^{-1}. Processing by ECAP by 8 passes reduced the grain size from an initial value of ~ 95 µm in the unprocessed condition to an equiaxed grain size of ~ 230 nm. The results show that the hardness and tensile strength are increased significantly after ECAP and the material exhibits excellent elongations up to ~12 % to 18 % in tensile testing at room temperature. The success in processing CP-Ti at room temperature with a 90° die provides opportunities for incorporating additional processing steps in order to gain further grain refinement and strength enhancement.

Key words*: equal channel angular pressing; titanium; mechanical properties; ultrafine grains*

Byl studován vývoj mikrostruktury a mechanických vlastností technicky čistého titanu jakosti Gr. 2 při aplikaci extrémní plastické deformace technologií ECAP. Použitá matrice ECAP měla vnitřní úhel β = 90°, protlačování bylo provedeno při teplotě 350 °C. Příčný průřez protlačovaných vzorků je 20 × 20 mm a délka vzorků 125 mm. Progresivní zařízení ECAP je instalováno v laboratoři RMVTC a je vybaveno protitlakem, který umožňuje zpracovávat i kovové materiály s velmi nízkými plastickými vlastnostmi.

V článku je detailně analyzován vztah mezi aplikovaným postupem ECAP a vývojem mikrostruktury a základních mechanických vlastností titanu v závislosti na velikosti aplikované deformace. Při protlačování titanu ECAP matricí probíhá už v počátečních průchodech matricí intenzivní zjemňování zrna. Se zvyšujícím se počtem průchodů matricí vzniká ve struktuře textura. Jednotlivá zrna jsou přednostně orientována ve směru hlavní deformace. Již v počátečních průchodech matrici se výrazně zvyšují pevnostní vlastnosti zkoumaného titanu, avšak výrazně klesají jeho plastické vlastnosti. Po osmi průchodech matricí dosahuje mez kluzu titanu hodnot kolem 680 MPa. Plastické vlastnosti se snižují přibližně na třetinu původní hodnoty, i když zjemnění zrna pokračuje. Pro zrovnoměrnění deformace v celém svém objemu byly vzorky po jednotlivých protlačeních pootočeny kolem podélné osy o 90° a znovu protlačovány. Při protlačování byly měřeny deformační síly, vypočítány deformační odpory a výpočtově určena deformační rychlost. Rozbor struktury byl proveden pomocí světelné mikroskopie a pomocí TEM. Mechanické vlastnosti vzorků po protlačování byly stanoveny zkouškou tahem a aplikací penetračního testu při teplotě místnosti.

Klíčová slova*: ECAP; titan; mikrostruktura; mechanické vlastnosti*

Ultrafine-grained materials were studied extensively in recent years for their new physical and enhanced mechanical properties. Ultrafine-grained materials were processed using various severe plastic deformation processing techniques, among which the most developed is the equal channel angular pressing (ECAP). The ECAP has been noted for its effectiveness in producing ultrafine-grained materials while maintaining their original geometry. In this technique, a sample is subjected to severe plastic deformation, as it is pressed through an angular die of two equal intersecting channels. For example, an effective strain is imposed on the sample by an ECAP die with a 90° channel-intersection angle (Fig. 1). Using this technique, ultrafine grains were obtained in various commercially pure titanium (CP Ti) and β titanium alloys for implants, e.g. Ti-13Nb-13Zr, Ti-15Mo-5Zr-3Al, Ti-30Ta, Ti-16Nb-10Hf, etc. [1 – 4]. Although further verification is still needed, the formation of subgrain boundaries from fine slip bands during the ECAP processing has been suggested to play a significant role in the grain refinement of those alloys.

ECAP processing of hexagonal closely packed metals, such as titanium, zirconium, hafnium, etc., is of great interest since twinning has been noted to play an important role in conventional plastic deformation of those alloys. Due to the fact that primary slip systems of hexagonal closely packed metals, on the other hand, are restricted to either basal or the prism planes in closely-packed directions (a-axes directions), i.e. in a slip, a variety of deformation modes of twinning combined with dislocation slips have been observed to take place in conventional deformation processes [5]. In the shear deformation by twining in hexagonal closely packed metals, atoms are displaced only in a fraction of an interatomic spacing. The maximum shear due to the twins in most of single crystalline hexagonal closely packed metals is therefore in the range from 0.15 to 0.25 [6], which is much smaller than imposed by each ECAP steps, which is ~ 1.80 in this study.

As commercially pure titanium has the low strength for practical applications, its strengthening by grain refinement using ECAP processing has been attempted for potential structural and biomaterial applications [7, 8]. It is of interest to refine the grains in an effective manner. It has been reported that ECAP route B_c, which rotates the workpiece by 90° clockwise between individual passes, is the most effective in refining grains for titanium [9]. It has been suggested that several factors, including deformation mode, have significant effects on the effectiveness of grain refinement. To study the grain refinement mechanisms and effectiveness of various ECAP routes, it is essential to first understand the deformation modes. Although it is known that both twinning and dislocation slips are necessary for the deformation of hexagonal closely packed metals, such as titanium, it is still unknown, how much each deformation mode contributes to the deformation. Deformation mechanisms of titanium during the deformation of titanium have not been studied.

In this paper, we attempt to analyse deformation mechanism of commercially pure polycrystalline titanium during ECAP processing. Commercially pure titanium was processed by one to eight ECAP passes.

Experimental material and procedures

A commercially pure titanium Gr. 2 was cut to the dimensions 20 × 20 mm and length of 125 mm and then annealed at 800°C for 1 h in Argon atmosphere, and then quenched in water of room temperature. Its chemical composition is shown in Tab. 1. Photograph of an ECAP die used by the RMSTC at the VŠB-Technical University of Ostrava (Fig. 2 and 3).

Tab. 1 Chemical composition of titanium Gr. 2 (wt.%)
Tab. 1 Chemické složení titanu Gr. 2 (hm.%)

C	Fe	H	O	N	Ti
0.06	0.02	0.012	0.22	0.03	~ 99,6

The average grain size was 95 μm after the heat treatment. ECAP was conducted using a die with an internal angle ϕ of 90° and an outer curvature angle Ψ of 20°.

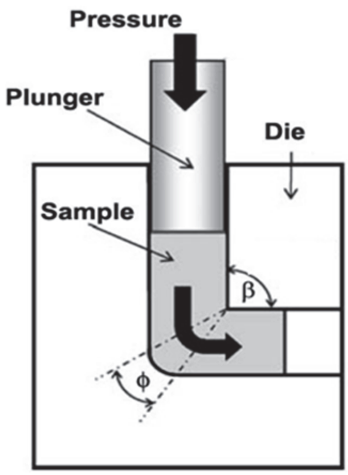

Fig.1 Schematic illustration of a die used in the present investigation, with β = 90° and ϕ = 20°
Obr. 1 Schema matrice a postup protlačování ECAP

Fig. 2 Photograph of an ECAP die used by the RMSTC at the VŠB-Technical University of Ostrava
Obr. 2 Zařízení pro aplikaci ECAP instalované v laboratořích RMVTC

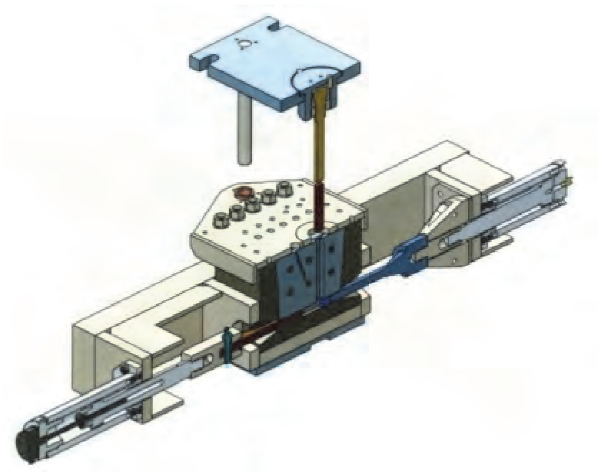

Fig. 3 ECAP die assembly fitted to the hydraulic press
Obr. 3 ECAP matrice instalovaná na hydraulickém lise

The present ECAP die was designed to give an approximate strain ε ~ 1 during each extrusion according to the following equation:

$$\varepsilon = \frac{N}{\sqrt{3}}\left[2cot\left(\frac{\beta}{2}+\frac{\phi}{2}\right)\phi\ cosec\left(\frac{\beta}{2}+\frac{\phi}{2}\right)\right] \quad (1)$$

where ε is the effective strain, N - number of passes, β - angle between the channels (inner arc angle), ϕ - outer arc angle

Repetitive extrusions of the same sample were performed in max. six passes. During the ECAP processing, all extrusions were conducted at the temperature 350 °C, using the procedure designated as the route B_c, in which each sample was rotated by 90° around its longitudinal axis between the individual passes. The details of the ECAP processing have been reported elsewhere [10]. Micro-hardness and tensile tests were carried out to evaluate the strength and ductility of the ECAP processed materials. Vickers micro-hardness (HV) was measured on the plane perpendicular axes, by imposing a load of 100 g for 15 s. Tensile properties in the transverse and longitudinal directions of the ECAP and non-ECAP Ti rods were measured using the miniature tensile specimens with a diameter of 3 mm and gauge length 15 mm (Fig. 4).

The tensile samples were extracted from the central portion of the ECAP and non-ECAP materials using electro-discharge machining. A displacement rate of 5 mm·min^{-1} was used for tensile testing, corresponding to an initial strain rate of 5.55×10^{-3} s^{-1}. The micro-structure of the ECAP samples was investigated using light microscopy and transmission electron microscopy (TEM).

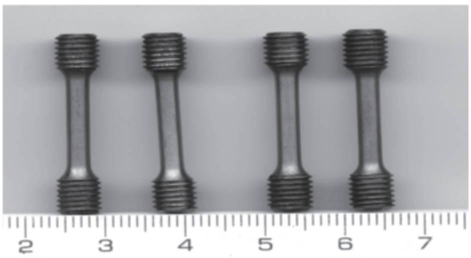

Fig. 4 Photographs showing the rods for tensile test
Obr. 4 Fotografie zkušebních vzorků pro tahovou zkoušku

The TEM specimens were thinned mechanically to a thickness of approx. 50 µm. Disks of 3 mm diameter were punched out from the thinned sheet, and polished by a jet polishing technique using a solution of 25% HF + 75% HNO_3. TEM observations were conducted using a JEOL JEM 2100 operated at 200 kV.

Results and discussion

All the samples were tested at room temperature by tensile test till rupture.

The engineering stress-strain curves of the initial samples (non-ECAP) and after ECAP processing of Ti from one pass to eight passes are shown in Fig. 5.

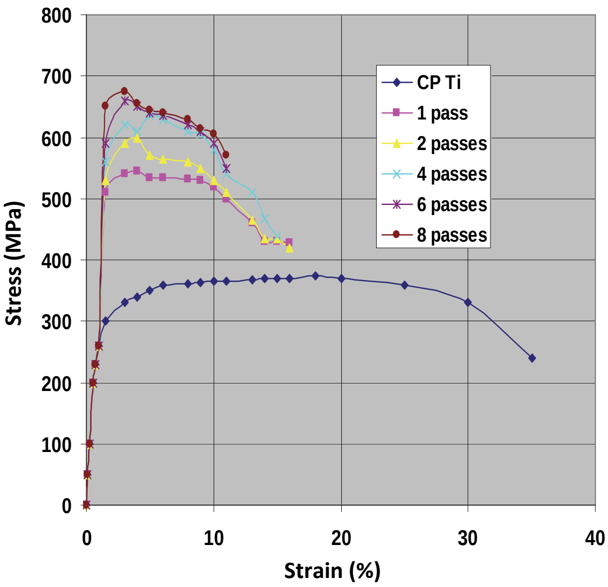

Fig. 5 Stress-strain curve for titanium Gr. 2 in initial state and after ECAP deformation
Obr. 5 Křivka napětí - deformace titanu Gr. 2 ve výchozím stavu a po deformaci ECAP

The data of yield stress (YS), ultimate tensile strength (UTS), hardness, elongation to failure, and grain size, are summarised in Tab. 2.

The first row in Tab.2 describes the properties in the initial state (prior to ECAP processing), and following rows give the properties in dependence on a number of passes through the ECAP tool. The columns from the second to the fifth give the following values: yield point, strength, ductility till rupture and micro-hardness, the last column expresses the average grain size.

Tab. 2 Mechanical properties of CP Ti before and after ECAP
Tab. 2 Mechanické vlastnosti CP Ti před a po ECAP

Passes	YS	UTS	El.	HV	ε	Grain size
	(MPa)		(%)	(-)	(-)	(µm)
0	300	385	38.4	156	0	95
1	500	550	18.3	199	1	2.15
2	540	605	18.2	205	2	0.52
4	580	630	17.9	210	4	0.35
6	605	685	12.4	240	6	0.26
8	680	690	12.2	251	8	0.23

The results confirm strengthening of CP Ti in dependence on the applied strain. The yield point increases from the initial value of 300 MPa to 680 MPa, and UTS increases from 385 MPa to 690 MPa after 8 passes through the ECAP tool. Application of severe plastic deformation is, however, manifested also by significant reduction of ductility of the samples achieving even 12 %, it means just to one-third of the initial value.

Deformation behaviour of ultra-fine grained titanium differs from the behaviour of titanium with ordinary grain size. After four passes through the ECAP tool, the fine-grained structure has increased the yield point from the initial value of 300 to 580 MPa. Similarly, the strength has also been increased, which increases rapidly till the fourth pass, after which the increase in strength is insignificant. The magnitude of real strain within the 4^{th} to 8^{th} passes increases the strength only by 60 MPa. In spite of that, it may be stated that technically pure titanium is a suitable material for biological applications. If we compare the increase in strength in coarse-grained and fine-grained materials, it is interesting that ultra-fine grained structure manifests decreasing intensity of strengthening in dependence on the magnitude of the applied strain. Proportionate changes of mechanical properties of CP Ti are given in Tab. 3.

Tab.3 Proportionate changes of mechanical properties of CP Ti and proportionate change of grain size (ΔGS) in dependence on the applied strain

Tab. 3 Poměrné změny mechanických vlastností CP Ti a poměrná změna velikosti zrna (ΔGS) v závislosti na aplikované deformaci

Passes	Δ YS	Δ UTS	Δ El.	ΔGS
	(%)			
Initial properties	300	385	38.4	95
	(MPa)		(%)	(μm)
ECAP 1	166.6	142.8	-52.4	226.3
ECAP 2	108.0	110.0	-0.55	24.2
ECAP 4	107.4	104.1	-1.65	67.3
ECAP 6	104.3	108.7	-30.7	74.2
ECAP 8	112.3	100.7	-1.62	7.69
$\Sigma\Delta x = (x_{i+1/}\ x_{i}).100$	226.7	179.2	-26.2	273.68

It may be assumed that strengthening of ultra-fine grained materials does not correspond to the classical Hall-Petch relation, which means that it is less sensitive to the change of grain size than it would correspond to ordinary (coarse-grained) structures [11].

Evolution of micro-structure of technically pure titanium is illustrated in Fig. 6.

More detailed picture of structure evolution was obtained by TEM. Figures 4 and 5 show photos of sub-structure in the initial state and after real (logarithmic) strain 8. Initial micro-structure of CP Ti was formed by equi-axed grains with a comparatively low density of dislocations. Fine particles of precipitates/inclusions were observed inside the grains [12]. A typical example of the structure is documented in Fig. 7.

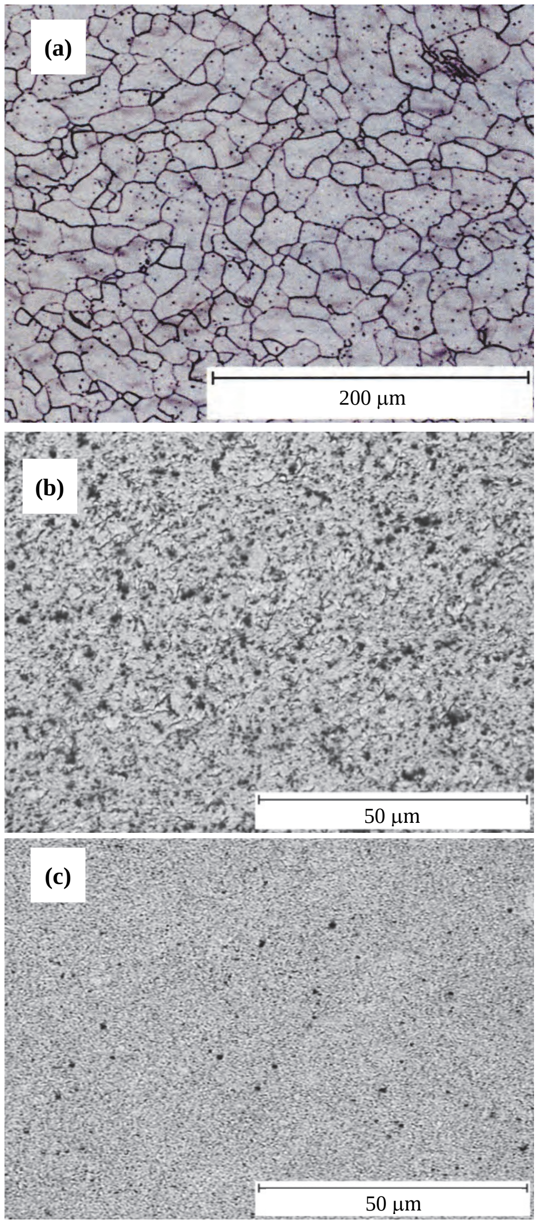

Fig. 6 Evolution of CP Ti micro-structure in dependence on magnitude of the applied strain: a) initial structure, b) e = 4, e = 8

Obr. 6 Vývoj mikrostruktury CP Ti v závislosti na velikosti aplikované deformace: a) počáteční struktura, b) e = 4, c) e = 8

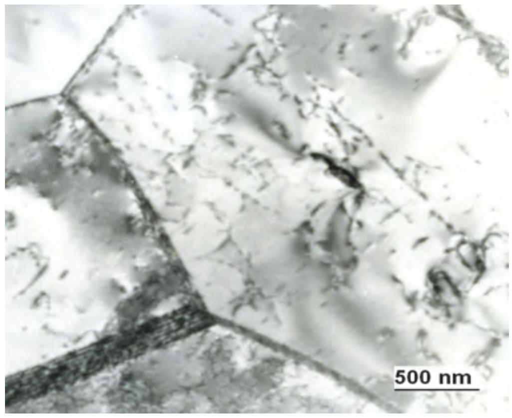

Fig. 7 Initial micro-structure of technically pure titanium

Obr. 7 Výchozí mikrostruktura technicky čistého titanu

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After 8 passes through the ECAP tool the structure was refined. Evolution of micro-structure of CP Ti in dependence on the magnitude of the applied strain is shown in Fig. 8.

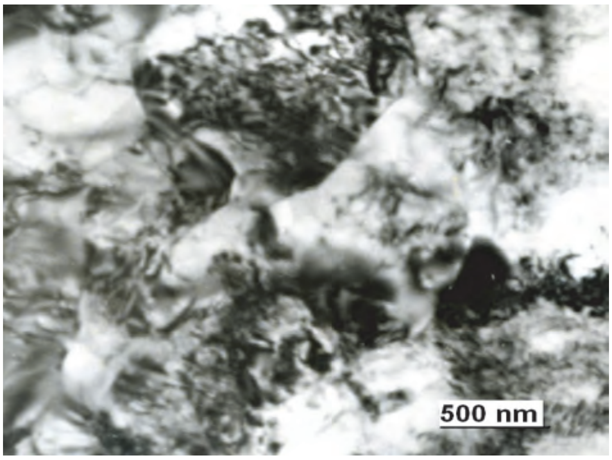

Fig. 8 Sub-structure of technically pure titanium after 8 passes through the ECAP tool

Obr. 8 Substruktura technicky čistého titanu po 8 průchodech ECAP

Grain size was non-uniform, apart from equi-axed grains the structure contained isolated areas of elongated, coarser grains (Fig. 9).

The fracture surface of CP Ti observed on the tensile test cross-section after 8 passes through the ECAP tool consists of individual pits, see Fig. 8. It may be assumed that comparatively big deformation energy was applied at the fracture, which was connected with the process of joining of individual fracture surfaces – cavities. The diameter of individual pits varies around 10 µm. Fracture character corresponds to a trans-crystalline failure.

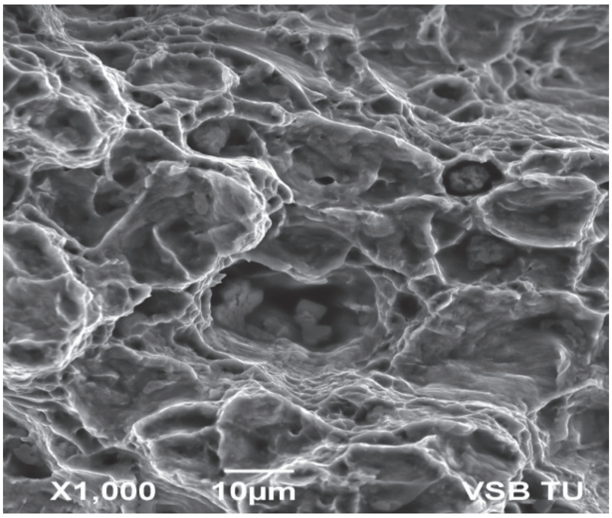

Fig. 9 Fracture surface of CP Ti after 8 passes through the ECAP tool

Obr. 9 Lomová plocha CP Ti po 8 průchodech ECAP

Conclusions

Technology for production of ultra-fine grained titanium was proposed and verified. Grain refinement of the initial CP titanium was achieved by the ECAP process and it depends on the magnitude of the applied strain. Strength properties of CP Ti were significantly enhanced by the grain refinement. Considering the comparatively low deformation temperature (350 °C) we assume, that the cause of strengthening may be both deformation strengthening, as well as strengthening as a result of grain size. We were unable to identify the share of individual strengthening mechanisms. Grain size after 8 passes through the ECAP tool varies around 230 nm and strength around 690 MPa.

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