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EFFECT OF SEVERE PLASTIC AND HEAVY COLD DEFORMATION ON THE STRUCTURAL AND MECHANICAL PROPERTIES OF COMMERCIALLY PURE TITANIUM

UČINEK PLASTIČNOSTI IN DEFORMACIJE PRI PODHLAJEVANJU NA STRUKTURNE IN MEHANSKE LASTNOSTI ČISTEGA KOMERCIALNEGA TITANA

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SPD (severe plastic deformation) processing of materials provides a great potential associated with the enhancement of their properties by refining the initial grain structure. The present experiments involved mechanical working of commercial-purity titanium (Ti Grade 2) with the CONFORM SPD technique, which is one of the SPD methods, and with rotary swaging. The objective was to process the material at as low temperatures as possible in order to avoid softening processes and, therefore, to achieve the maximum strengthening through a microstructure refinement. Three passes through a CONFORM SPD machine were completed and the resulting ultimate strength was 673 MPa. The average grain size was 330 nm. The greatest improvement of the mechanical properties was achieved in the first pass. In the subsequent passes, the contributions were minor. The processing in the CONFORM SPD machine did not impair the ductility of the material. Subsequently, the wires were rotary swaged. The ultimate strength achieved was 1070 MPa. The response of the properties to this forming method was markedly different. The reason is that rotary swaging does not belong to SPD techniques. It causes rapid work hardening and reduces the ductility of the material. The workpiece was subsequently investigated with the aid of several techniques. Light and transmission electron microscopy and X-ray diffraction were employed for evaluating the grain size, distribution and orientation.

Keywords: equal-channel angular pressing, CONFORM SPD technique, rotary swaging, titanium, extrusion

Velika plastična deformacija (angl. SPD) pri obdelavi materialov omogoča veliko možnosti, povezane z izboljšanjem njihovih lastnosti z izboljšavo izvorne strukture zrn. Pričujoči preizkus je vključeval mehansko obdelavo komercialno dostopnega čistega titana (Ti Grade 2) s CONFORM SPD-tehniko, ki je ena od SPD-metod, in s kovanjem. Namen je bil, da bi se material obdelalo pri čim nižjih temperaturah, kot je mogoče, da bi se izognili procesom mehčanja in, da bi dosegli maksimalno krepitev s prečiščenjem strukture. Narejeni so bili trije nizi v CONFORM SPD-stroju in končna moč je dosegla 673 MPa. Povprečna velikost zrn je bila 330 nm. Največje izboljšave mehanskih lastnosti so bile dosežene v prvem nizu. V sledečih nizih so bile le-te manjše. Obdelava v CONFORM SPD-stroju ni škodovala duktilnosti materiala. Posledično so bile žice zvite. Končna moč je bila 1070 MPa. Odziv lastnosti na to metodo oblikovanja je bil izrazito drugačen. Razlog je v tem, da rotacijsko zvijanje ne sodi v tehnike SPD. Rotacijsko zvijanje povzroči hitro utrjevanje in zmanjšuje duktilnost materiala. Vzorec je bil nato raziskan s pomočjo več tehnik. Za ovrednotenje velikosti, razporeditve in orientacije zrn, sta bili uporabljeni transmisijska elektronska mikroskopija in rentgenska difrakcija.

Ključne besede: enakomerno kotno stiskanje, CONFORM SPD-tehnika, rotacijsko zvijanje, titan, iztiskanje

1 INTRODUCTION

Severe plastic deformation (SPD) is a generic term for a group of methods which cause grain refinement in a material and produce equiaxed grains.[1] The occurrence of an ultrafine structure or nanostructure is conditional on high hydrostatic pressure (P >1 GPa) and large shear deformation applied at relatively low temperatures. The temperatures of SPD processes should meet the condition of T(SPD) < 0.4 T(melting).[1] Another aspect is the strain magnitude, defined as e(true strain) > 6–8. When the above conditions are met, the forming process leads to a high density of lattice defects, predominantly dislocations, and to the formation of subgrains, which reduces the stored energy.[2] The relationship between the mechanical properties and the grain size is described with the Hall-Petch equation[3,4]. In the present study, commercially pure (CP) titanium Grade 2 was worked by means of the CONFORM SPD (CONSPD) method. This method uses continuous angular pressing through a die chamber of a modified design to produce ultrafine structures and nanostructures of the materials.[5–7] The design modification reflects the principles of the ECAP method (equal-channel angular pressing), as described in studies.[5–7] Continuous operation is the key advantage of this forming method. The processing of CP titanium Grade 2 with the CONFORM SDP machine was described in multiple papers.[5,7] The main outcomes were the improved mechanical properties (ultimate tensile strength (UTS) = 698 MPa, 0.2 % offset yield stress (0.2 OYS) = 637 MPa), no significant decrease in the elongation, and an equiaxed ultrafine-grained (UFG) microstructure.[7]

The present study proposes a process route which involves CONFORM SPD forming and rotary swaging (RS) in order to enhance the mechanical properties of a workpiece.[8–10] The effects of severe plastic deformation and work hardening are thus combined. The proposed technology allows the mechanical properties of pure titanium to be improved dramatically. The ultrafine-grained CP Ti Grade 2 has a great potential because its mechanical properties are comparable to the Ti6Al4V alloy, which is mostly used in medicine. Recently, it was suggested that Al and V might be toxic for the human body.[11,12]

2 EXPERIMENTAL PART

The material under investigation was CP titanium Grade 2 with the chemical composition given in **Table 1**. The composition was measured by means of the Bruker Q4 Tasman optical emission spectrometer and the Bruker G8 Galileo gas analyser. The diameter of Ti rods was 10 mm.

Table 1: Chemical composition of feedstock in weight percent

Fe	O	C	H	N	Ti
0.046	0.12	0.023	0.0026	0.0076	99.822

The processing was carried out in a CONFORM SPD machine, type 315i with a modified die chamber (**Figure 1**), and in a HMP R4-4 rotary-swaging machine. During the CONFORM SPD process, the temperature was 220 °C, the wheel speed was 0.5 min^{-1}, and the angle of the die chamber was 90°. Three passes through the CONFORM SPD machine were completed. The product's cross-section was identical to that of the feedstock. Rotary swaging, the subsequent process, was carried out at ambient temperature. In this operation, the cross-section area was reduced by 20 % in each pass. The total area reduction was 90 %.

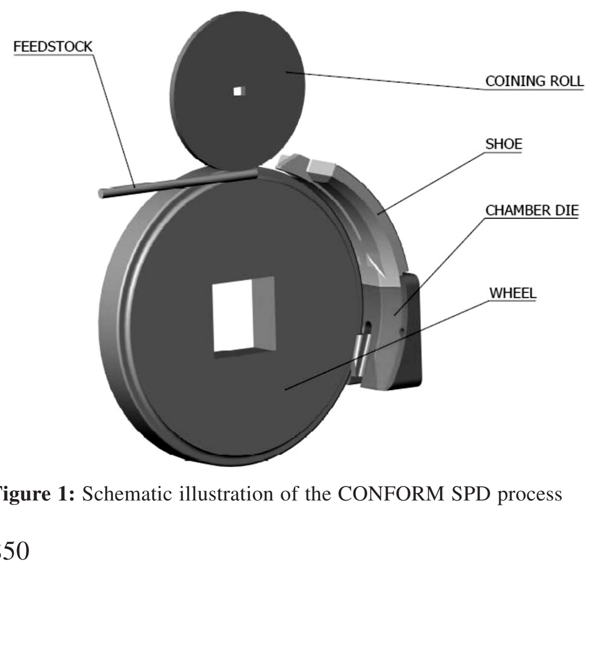

Figure 1: Schematic illustration of the CONFORM SPD process

For the purpose of observation with a transmission electron microscope (TEM), thin foils were prepared with the final electrolytic thinning in a Tenupol 5 device, using a solution of 300 mL CH_3OH + 175 mL 2-butanol + 30 mL $HClO_4$ at –10 °C and a voltage of 40 V. The TEM analysis was performed with a JEOL 200CX instrument with an acceleration voltage of 200 kV. Selective electron diffraction was used for the determination of the phases. The grain size was measured using the linear intercept method.

The preferred orientation of crystallites (texture) was analysed with an automatic powder diffractometer X'Pert-Pro equipped with an ultra-fast semiconductor position-sensitive detector Pixcel. Cu-$K_{\alpha 1}$ radiation (λ = 0.154056 nm) was used. The texture was characterized from the radial X-ray diffraction patterns using the Harris (1952) texture index, Equation (1):

$$T_i = \frac{n \cdot I_i / R_i}{\sum_{j=1}^{n} I_j / R_j} \quad (1)$$

where n is the number of the reflections investigated, Ii is the observed intensity and R_i is the corresponding intensity for the sample with randomly oriented crystallites. The R_i values were obtained from the ICDD PDF standard diffraction data file (reference code 00-005-0682). The texture was analysed in the direction of the material flow, both after CONFORM SPD (longitudinal direction), and after CONFORM SPD + rotary swaging.

3 MICROSTRUCTURE

The mean grain size in the initial condition of the feedstock was 5390 nm. After the first pass, the mean grain size was d_{average} ~ 320±35 nm in the transverse direction. After the first pass, the microstructure was equiaxed with a non-uniform dislocation density (**Figure 2a**). Some locations exhibited a preferential orientation (**Figure 2b**). After the second pass (**Figure 2c**), the mean grain size was d_{average} ~ 250±25 nm in transverse direction. The dislocation density was still non-uniform after the second pass. After the third pass, the mean grain size was d_{average} ~ 330±30 nm (**Figure 3d**): the grain was larger than after the second pass. This increase can be attributed, in part, to the non-uniform deformation and to the high surface activity of the UFG microstructure. High surface activity and dislocation density (strain magnitude) lower the temperatures of softening processes. A certain grain growth can be expected to occur during the forming due to deformation heat and the heat retained in the die chamber (220 °C). The grain growth upon multiple passes through the CONFORM SPD machine was already reported in [9].

Figure 3a is a micrograph of the structure in the transverse direction after three passes through the CONFORM SPD machine and after a 35 % area reduction

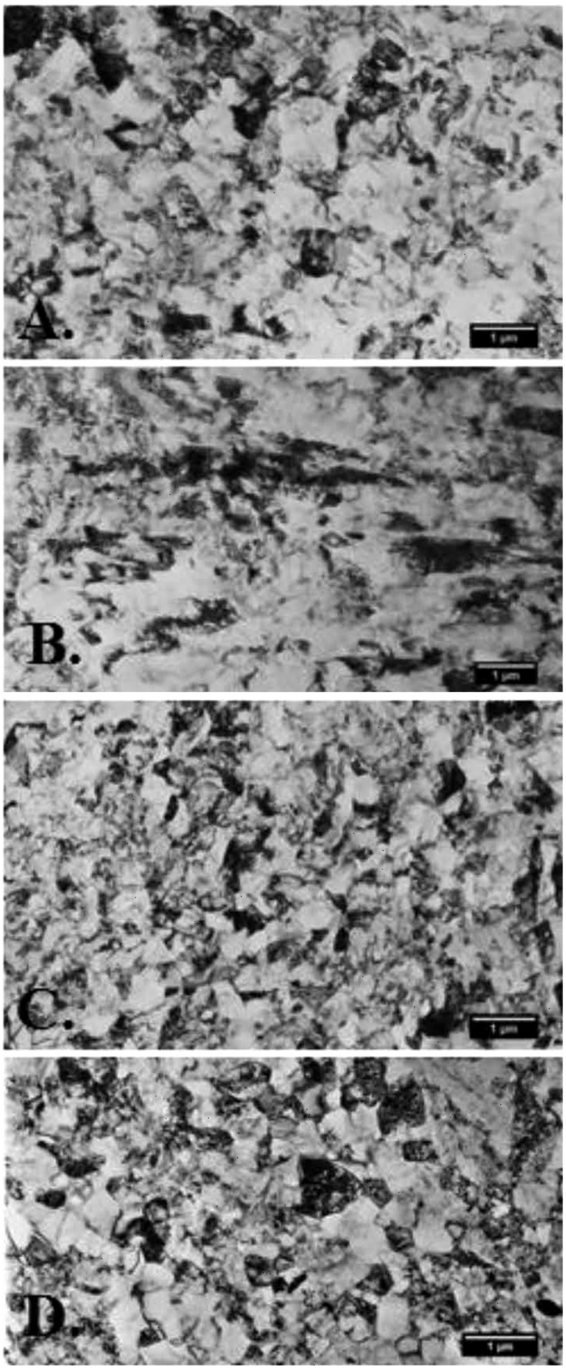

Figure 2: TEM images: a) CON SPD – 1 pass (transverse), b) CON SPD – 1 pass (longitudinal), c) CON SPD – 2 passes (transverse), d) CON SPD – 3 passes (transverse)

with rotary swaging (RS). The mean grain size in the transverse direction is d_{average} ~ 300±150 nm. The large standard deviation is due to a large variation in the grain size. **Figure 3b** is a micrograph taken in the longitudinal direction after CONFORM SPD and RS. The grains are elongated and aligned in the direction of forming due to the nature of the deformation introduced by rotary swaging. Unlike in CONFORM SPD, the grains become elongated instead of being refined or converted into new polyhedral grains. After the additional reduction due to rotary swaging (3 passes through CONFORM SPD + the total area reduction of 50 % by RS), it became impossible to determine the grain size due to the high dislocation density, as seen in **Figure 3c**. The summary of the average grain sizes is given in **Table 2**.

Table 2: Average grain size in the transverse and longitudinal sections obtained from TEM images

Condition	Grain size (nm)	
	Transverse	Longitudinal
as received	5390±20	
CON SPD – 1 pass	320±35	340±30
CONSPD – 2 passes	250±25	310±30
CONSPD – 3 passes	330±30	420±30
CON SPD – 3 passes + RS (35 %)	300±150	220±50
CONSPD – 3 passes + RS (50 %)	Not possible to evaluate due to high dislocation density	

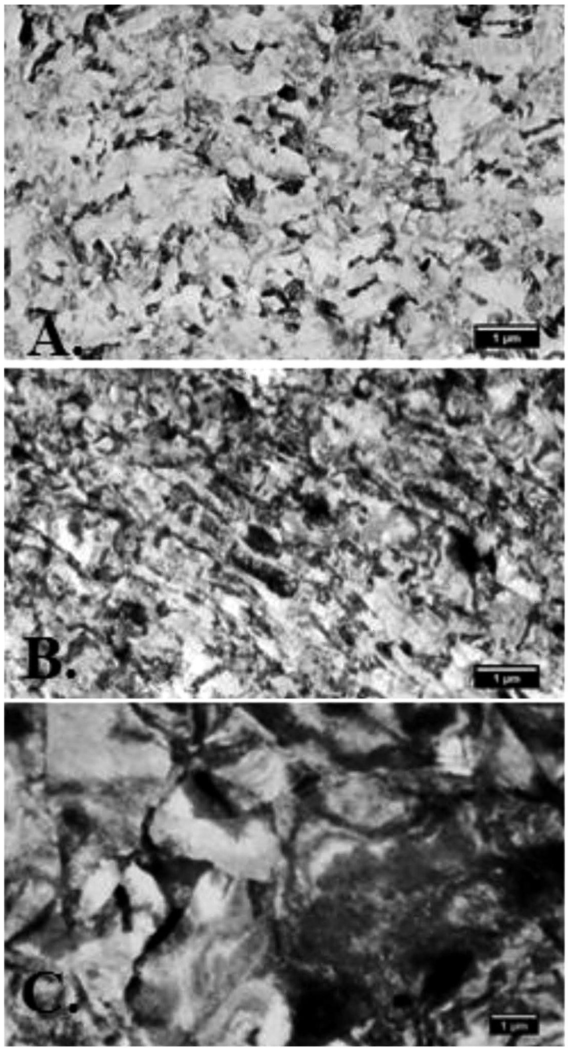

Figure 3: TEM images: a) CON SPD – 3 passes + RS (area reduction by 35 %) in the transverse direction, b) CON SPD – 3 passes + RS (35 %) in the longitudinal direction, c) CON SPD – 3 passes + RS (50 %) in the transverse direction

4 TEXTURE EVALUATION

The preferred orientation of crystallites expressed with the texture index Ti (Equation 1, **Table 3**) is, in all the cases, in the [001] direction perpendicular to the sample surface analysed. The highest value was found for CONSPD – 3 passes + RS (75 %) titanium sample, i.e., the sample that was processed using the CONFORM SPD device and rotary swaging (basal texture, **Figure 5**). An increased value of the texture index can be seen in the initial state, due to the previous wire-drawing process (**Figure 4**). The texture index is lower for the samples that were processed with the CONFORM SPD device. Texture indices for the [101] direction are listed for the sake of comparison. Nevertheless, all their values are lower than 1. It means that no preferred orientation of crystallites in the [101] direction is present, as opposed to the sample surface.

Table 3: Texture evaluation using X-ray diffraction

Plane (h k l)	Texture index		
	as received	CONSPD – 1 pass	CONSPD – 2 passes
$T_{i\,(002)}$	3.13	2.49	2.67
$T_{i\,(101)}$	0.66	0.69	0.86
	CONSPD – 3 passes	CONSPD – 3 passes + RS (75 %)	Standard
$T_{i\,(002)}$	2.37	3.48	1
$T_{i\,(101)}$	0.97	0.24	1

4 MECHANICAL PROPERTIES

Table 4 indicates that the largest increase in the mechanical properties was obtained during the first pass through CONFORM SPD. The subsequent passes led to smaller increments. It is important to note that the increase in the ultimate strength and yield stress upon CONFORM SPD is not offset by a decrease in the ductility. The improvement in the mechanical properties is mainly due to the grain refinement and the increased

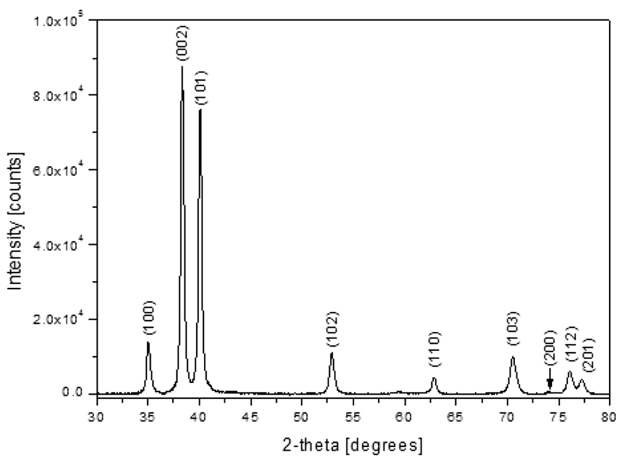

Figure 4: Diffraction radial profile of the as-received specimen

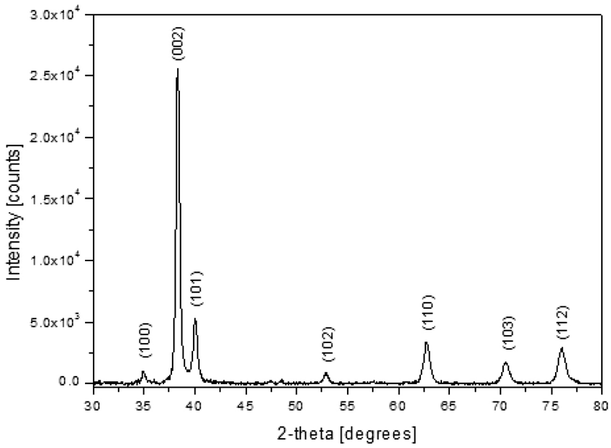

Figure 5: Diffraction radial profile of the specimen with the strongest texture: CONSPD – 3 passes + rotary swaging (75 %)

dislocation density, which impede the dislocation movement. A further increase in the mechanical properties was obtained with rotary swaging applied after the three passes through the CONFORM SPD machine. In comparison with the CONFORM SPD process, rotary swaging leads to a decrease in the ductility as a result of work hardening.

Table 4: Mechanical properties in the as-received condition, after CONFORM SPD processing and after RS

Condition	0.2 OYS	UTS	A_5	RA
	MPa	MPa	%	%
as received	370	480	25	52
CONSPD – 1 pass	540	580	23	62
CONSPD – 2 passes	560	600	23	62
CONSPD – 3 passes	570	623	20	64
CONSPD – 3 passes + RS (50 %)	830	885	13	54
CONSPD – 3 passes + RS (75 %)	930	1000	12	57
CONSPD – 3 passes + RS (85%)	950	1070	12	58
RS only (85 %)	780	850	12	50

5 DISCUSSION

The CONFORM SPD processing led to higher ultimate strengths and yield stresses without a reduced ductility. This is characteristic of the process of the formation of a UFG equiaxed structure shown in **Figure 2**. The grain size and distortion are non-uniform due to non-uniform deformation during the forming process. The preferred orientation of crystallites in the [001] direction perpendicular to the surfaces of the investigated samples is not too significant, except for the sample which was processed with CONFORM SPD and the rotary swaging machine where the highest value of the texture index was observed (the basal texture).

Major increases in the ultimate strength and yield stress were found after the first pass through the CON-

FORM SPD machine, whereas the increments from the subsequent passes were small. This is closely related to the grain size, which did not decrease during the subsequent passes. It is possible to say that the additional deformation causes the subgrains to rotate into high-angle grain boundaries, typically with an equiaxed shape of the grains.[5,7,9] The subsequent rotary swaging led to a further increase in the mechanical properties, but the strengthening mechanism was different this time. Instead of the formation of equiaxed grains, the grains became elongated in the direction of forming, with a much higher dislocation density (**Figure 3**).[10,11] When compared to the CONFORM SPD route, rotary swaging led to a reduced ductility as a consequence of work hardening.

6 CONCLUSION

This study involved the processing of Ti Grade 2 using CONFORM SPD and rotary swaging with the goal of improving its mechanical properties. The results and findings are described below:

- The CONFORM SPD processing substantially refined the grain from its initial size of 5390 nm to 350 nm. The subsequent rotary swaging did not provide for a further grain refinement but led to a grain elongation and to a higher dislocation density.
- The processing led to the basal texture. The most significant form of texture was found in the specimen processed with CONFORM SPD and rotary swaging.
- Three passes through the CONFORM SPD machine led to a strength of 623 MPa and a yield stress of 570 MPa without any notable decrease in the elongation.
- After CONFORM SPD, the specimens were rotary swaged, which brought their strength to 1070 MPa and the yield stress to 950 MPa. In the initial condition, the ultimate strength was 480 MPa and the yield stress was 370 MPa.

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