

# Materials development on the nanoscale by Accumulative Roll Bonding procedure

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The paper presents some scientific results referring to Commercially Ti Grade 2 processed by SPD (Severe Plastic Deformation) to reach nanometric scale. Accumulative Roll Bonding (ARB) procedure offers the possibility to refine the grain in bulk material and therefore the sensible improvement of mechanical properties of the metallic materials utilized in practical applications. The scientific research is focused on ARB process settlement. Also, structural analysis and mechanical tests were made on processed material. The results regarding the possibility to obtain structures in UFG (Ultra Fine Grain) range or NC (Nanocrystalline Grains) range confirm that, in the case of Commercial Ti Grade 2, the use of SPD techniques, more precisely ARB (Accumulative Roll Bonding), leads to structures placed in UFG domain (0.780  $\mu\text{m}$ ).

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## 1. Introduction

The development of metallic materials with grain dimensions on the nanoscale represent a research area of great interest in the last years.

That interest increases thanks to the attractive properties of this type of materials, such as: high mechanical resistance, high toughness and good wear resistance.

Several processing techniques [1] are currently available to obtain ultra-fine grained or nano-crystalline grains in different materials types and they can be classified in the following four groups:

- mechanical technique – that includes cryo-milling and compaction;
- severe plastic deformation, SPD;
- gas-phase condensation of particulates and condensation;
- electrodeposition;

The SPD techniques offer the possibility to refine the grain in bulk material and therefore the sensible improvement of mechanical properties of the metallic materials utilized in practical applications. The most important SPD procedures are:

- severe Plastic Torsion Strain – SPTS;
- high Pressure Torsion – HPT;
- cyclic Channel Die Compression – CCDC;
- friction Stir Processing – FSP;
- equal Channel Angular Pressing – ECAP;
- accumulative Roll Bonding – ARB;
- cyclic Extrusion and Compression – CEC

The SPD procedures lead to a great variety of materials processed, to advanced chemical composition purity and high mechanical proprieties.

Using these techniques have been obtained ultra fine and nanocrystalline structures for different types of metallic

materials like: Fe-Armco (ECAP), titanium alloys, titanium with different grade of purity (ECAP), IF iron (ARB), aluminum base alloys, AZ91, (ARB) [2,3].

From all SPD techniques, most known method is ECAP. The ECAP procedure involves deforming a billet of material by shear without changing its overall dimensions.

As a SPD method, ARB involve severe deformation of metal sheets without modification of overall material thickness.

The technique consist in rolling of two metal sheets with equal dimensions using a 50 % percent reduction until sheet that result has the same thickness with the originals sheets.

Using the ARB process is possible to achieve extremely high plastic deformation of material, because theoretically, the number of cycles can be repeated without a limit. At each ARB step the one single layer can support an added deformation up to theoretical limit.

The surfaces in contact during rolling process must submit a mechanical or chemical cleaning before ARB procedure, because it is necessary to remove impurities and oxide layers. This cleaning assures a certain roughness which is helpful to realize adhesion between sheets. After each ARB step, the rolled material is cut in two equal pieces, stacked together and rolled again.

The process is repeated until the desired deformation degree is reached. The obtained deformation is practically unlimited because the ARB steps can be repeated without a limit.

An important aspect of ARB procedure is the layer bonding process. The adhesion depends strongly not only on the surface treatment and rolling temperature, but also depends on initial sample geometry. The critical parameters referring to rolling temperature and initial material dimensions must be settled for each material case.

Using the ARB technique, good results were obtained in case of certain metallic materials like: IF iron (%C=0.0031, %Si< 0.01, %Mn=0.15; %P=0.01; %S=0.005; %Ti=0.049; %B< 0.0001; %sol Al<0.054; %Fe=bal)[4],[5].

## 2. Experimental

It is known that titanium has a large palette of applications in different technical areas and medical field.

The experiments are focused to obtain ultrafine or nano structured commercially Grade 2 titanium, using ARB technique, because ARB technique is not so expensive and it is relative simple.

The titanium Grade 2 samples used in experiments were roll-milled using a laboratory roll-mill which has the following parameters:

- cylinder diameter:  $\phi$  150 mm;
- cylinder length: 300 mm;
- rolling mill power: 45 kW;
- rolling mill speed: 1.5 m/s.

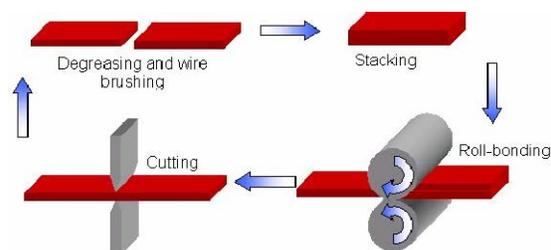


Fig. 1. Schematic representation of ARB procedure.

The initial sample used to initiate the ARB cycle had an initial diameter of 3 mm; this one was roll-milled to h x l: 0.8 x 9.6 mm. All rolling steps were performed after the samples were heated at 800 °C in a CALORIS laboratory electrical oven.

After each rolling step the samples were mechanical cleaned in order to eliminate oxide layers and other impurities.

The performed ARB procedure consist in 10 rolling cycles, as shown in table 1.

Table 1. Obtained results during ARB procedure.

No. of cycles		1	2	3	4	5	6	7	8	9	10
No. of layers		2	4	8	16	32	64	128	256	512	1024
Stacke thickness [mm]	before deformation	1.6	1.6	1.6	1.6	1.6	1.6	1.6	1.6	1.6	1.6
	after deformation	0.8	0.8	0.8	0.8	0.8	0.8	0.8	0.8	0.8	0.8
Layer thickness* [mm]	before deformation	0.8	0.4	0.2	0.1	0.05	0.025	0.0125	0.00625	0.00312	0.00156
	after deformation	0.4	0.2	0.1	0.05	0.025	0.0125	0.00625	0.00312	0.00156	0.00078
Partially deformation degree, $\epsilon$		0.5	0.75	0.875	0.9375	0.9687	0.9843	0.9921	0.996	0.998	0.999
Medium width** [mm]		11.3	14.7	18.1	23.4	25.2	26	27.2	ct.	ct.	ct.

\* the layer thickness was theoretically calculated by dividing the sample stack thickness at the number of layers contained in that stack;

\*\* at the end of ARB procedure, material presents important irregularities, the medium width of the stack can't be exactly measured, it can be only appreciated as being constant.

## 3. Results

The tensile tests were performed in traction mode using an INSTRON 3380 Universal Testing Machine. The tests parameters were:

- testing temperature: 20 °C;
- crosshead speed for traction tests: 2.54 mm/min;

Obtained Strain-Stress curve profiles are shown in figure 2, 3 and 4.

Structural analysis was made by optical and SEM microscopy. For optical microscopy were used samples retained after 7, 8 and 10 ARB cycles, in order to reveal the structural aspects. The samples were prepared and

etched using a HF - HNO<sub>3</sub> - H<sub>2</sub>O solution. Obtained micrographs are presented in figure 5, 6 and 7.

To a more detailed investigation about samples microstructure, SEM investigations were made on samples with 128, 256, 512 and 1024 layers. Significant SEM image are presented in figure 8 and 9.

## 4. Discussion

The tensile tests were performed on samples retained in early stages of ARB procedure. In these stages the overall sample geometry is more suitable to tensile tests. At the end of ARB procedure the general samples shape becomes irregular, which can be an inconvenient in tensile

tests accuracy. The irregularities which appear at each ARB step are also sources for samples premature fracture (fig. 4). The strain-stress curves show that material strain decrease with increasing number of ARB cycles. No relevant stress increasing was observed on samples tested.

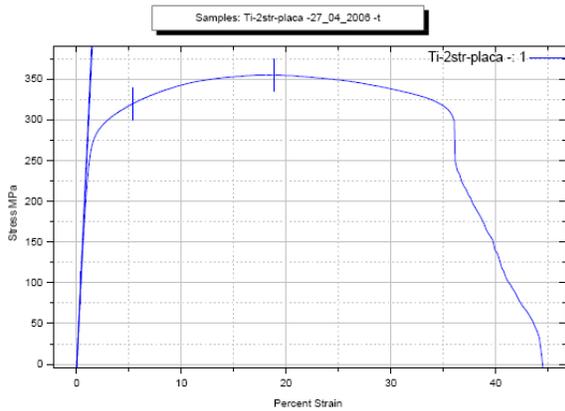


Fig. 2. Strain – Stress curve profile for sample with 2 layers.

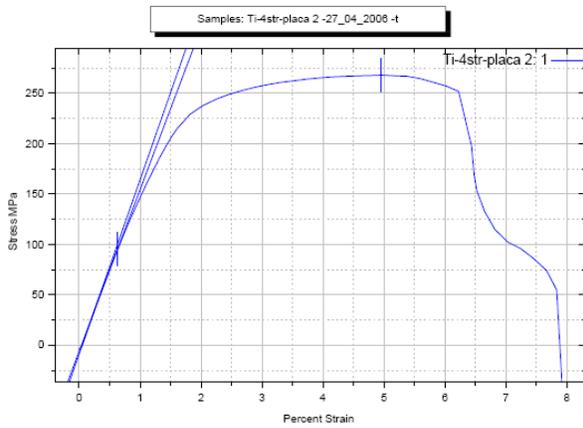


Fig. 3. Strain – Stress curve profile for sample with 4 layers.

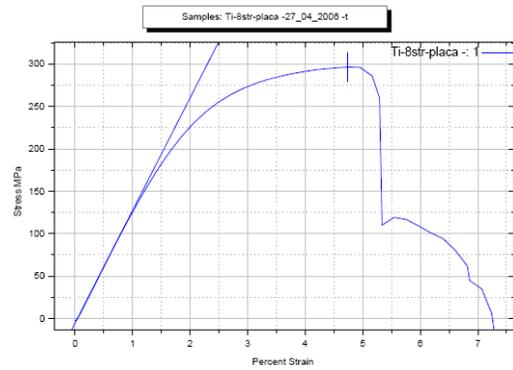


Fig. 4. Strain – Stress curve profile for sample with 8 layers.

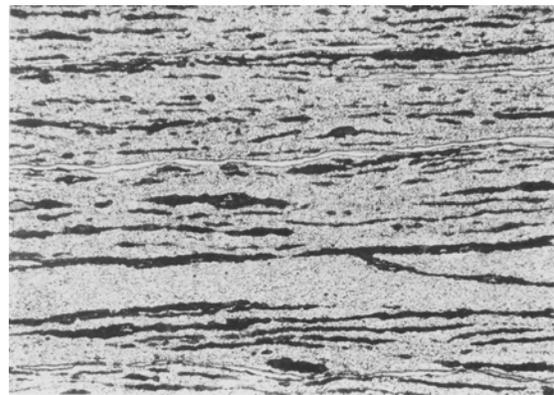


Fig. 5. Optical micrograph for sample with 128 layers; 100:1.

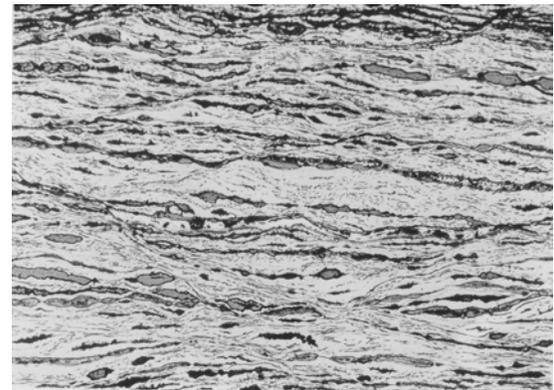


Fig. 6. Optical micrograph for sample with 512 layers; 100:1.

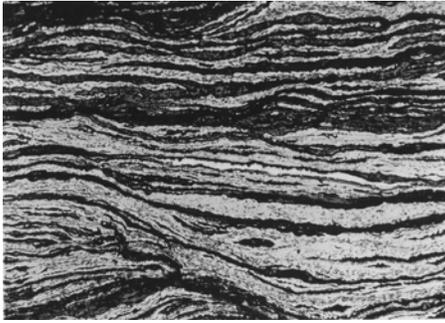


Fig. 7. Optical micrograph for sample with 1024 layers; 100:1.

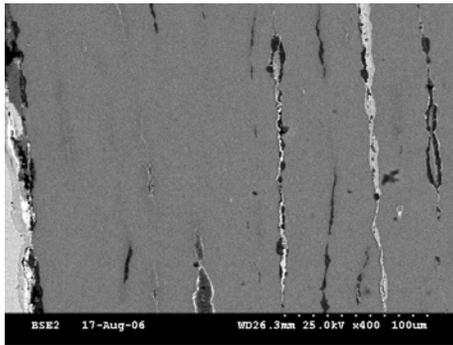


Fig. 8. SEM image for sample with 128 layers.

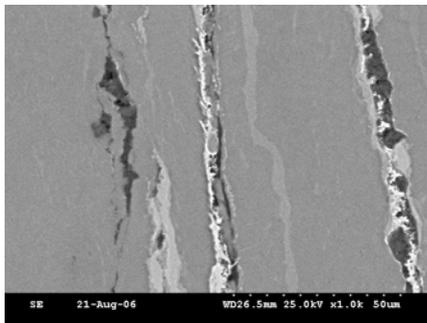


Fig. 9. SEM image for sample with 1024 layers.

The optical microscopy was performed in order to observe the general geometry of layers in rolled samples. One can see in Figs. 5 and 6 the presence of small grains inside layers, which can't be further detected in micrograph shown in Fig. 7, for the sample at the end of ARB procedure. All micrographs show some discontinuities and also impurities and oxide retention between layers. The degree of deformation non-uniformity increases with number of layers, but it is an intrinsic attribute of rolling process; it can be attenuated if one uses appropriate mill cylinder geometry and well equipped rolling stand. The SEM images show also the presence of impurities and oxides.

## 5. Conclusions

It is possible to obtain ultra fine (UFG) and nanocrystalline (NC) grains for commercially Grade II Titanium by ARB process;

From experimental data the estimated layer thickness is  $0.781 \mu\text{m}$  after 10 rolling cycles. This result may presume that the real grain size inside one layer is smaller and possible in nanometric scale.

The ARB process must be done in more accurate conditions. During rolling stages different inconvenient may occur, such as: im-purification between layers (contamination with other materials); local micro-fractures (affecting sample macro-geometry); local deformation non-uniformities (related to rolling equipment conditions). These inconvenient have an negative influence upon material mechanical properties. Generally speaking the material resistance is higher with increasing number of layers but it is possible a premature fracture due to deformation non-uniformities and to micro-fractures induced by ARB process. The strain – stress curve profile shows those influences.

All those inconvenients can be easily eliminated by an adequate working procedure.

In order to obtain adequate results using ARB process to obtain UFG and NC structured materials more investigations need to be done regarding:

- rolling temperature for an easy bonding between layers;
- the thickness of initial sheet and stack geometry;
- methods used in early stages of process to bond the stack;
- investigations upon microstructural grain transformations inside single layer and between layers which is different from one material type to other.

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## References

- [1] K. S. Kumar, H. Van Swygenhoven, S. Suresh, *Acta Mater.* **51**, 5743 (2003).
- [2] L. J. Slokar, T. Matkovic, P. Matkovic, *Metalurgija* **43**(4), 273 (2004).
- [3] D. Raabe, M. Sachtleber, Z. Zhao, *Acta Mater.* **49**, 3433 (2001).
- [4] R. Kocich, M. Greger, *Acta Metallurgica Slovaca* **11**(3), 277 (2005).
- [5] M. Haouaoui, I. Karaman, H. J. Maier, K. T. Hartwig, *Met. Mat. Trans. A* **35A**, 2935 (2004).

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