Abstract

The possibility of mass production of ultrafine-grained (UFG) titanium by industrial caliber rolling was examined. The mechanical and metallurgical characteristics of the rolled materials were studied. The process led to an UFG microstructure with high dislocation density, accompanied by high tensile strength and good ductility. Mathematical modelling indicated that the refinement of the grains was caused by the large shear strains and the non-monotonicity of deformation, i.e. caliber rolling can be regarded as a severe plastic deformation procedure. In the model, the cylindrical cavities and the other machine elements were assumed to be perfectly rigid, while the deformed material was assumed to be elastic-plastic. Temperature dependence of the material parameters was taken into account. From the model calculations the cooling time between each cavity, and the cavity filling were determined too. In addition, the characteristics of the samples processed by industrial caliber rolling were compared to those produced under laboratory conditions. The microstructure and the mechanical properties of the rolled materials indicate that industrial caliber rolling is capable of mass production of UFG titanium.

Keywords

Caliber rolling, ultrafine-grained material, titanium, non-monotonicity of deformation, finite element modelling

1. Introduction

In the past decades, several severe plastic deformation (SPD) procedures were developed for processing bulk metallic materials with ultrafine-grained (UFG) microstructure. Accumulative roll bonding (ARB) [1,2], equal channel angular pressing (ECAP) [3,4], high pressure torsion (HPT) [5] and repetitive corrugation and straightening [6] are frequently applied SPD techniques. SPD methods usually produce UFG materials under laboratory conditions, i.e. their productivity is low. Significant breakthrough in the application of UFG materials (i.e. their commercialization) can only be achieved if they were manufactured in an industrial environment.

The common characteristic of the SPD techniques is the large shear strain and the non-monotonic nature of the deformation. The concept of monotonic deformation was introduced by Smirnov-Aljajev [7]. As he wrote, a forming process develops monotonically if no component of the rate of strain tensor changes its sign, i.e. the eigenvectors of the rate of strain

1 Budapest University of Technology and Economics, Department of Materials Science and Engineering, Budapest, Hungary
2 Bay Zoltán Nonprofit Ltd for Applied Research, Engineering Division, Miskolc, Hungary
3 University of Dunaújváros, Dunaújváros, Hungary
4 Eötvös Loránd University, Department of Materials Physics, Budapest, Hungary
tensor are parallel to the eigenvectors of the strain tensor during the whole deformation process and the Lode parameter remains constant. Studying the possible ways of deviation from the monotonic deformation contributes to the development of processes which assure the production of fine grains. Although caliber rolling is not a usual SPD method, its industrial application for producing UFG titanium is promising due to its productivity and non-monotonic nature.

According to former experience, titanium is an ideal material for devices to be implanted into the human body. It is chemically inert, does not react with human body fluids, and, if it is necessary, after proper surface treatment the bone is able to adhere to titanium implants. However, if the adherence of the implant to the bone is contraindicated, it can be impeded by tailoring the titanium surface morphology. This dual feature makes this metal well and widely applicable for different medical purposes.

2. Rolling in industry

Grade 2 titanium specimens with 1000 mm in length and 70 mm in diameter in an annealed condition (annealing at 650 °C for 2 hours, then cooling in air) were used for the rolling tests. The initial mechanical properties were as follows: yield strength, YS = 332 MPa, ultimate tensile strength, UTS = 439 MPa, reduction in area, Z = 58 %, elongation to failure, A = 22 %, strain energy density to fracture, W_f = 412 J/cm^3.

Industrial warm caliber rolling of titanium was performed using the rolling mill system of OAM Ózd Steelworks Ltd. (Ózd, Hungary). The shape and the size of the roll cavities for the twelve passes are shown in Fig. 1 and Table 1. At the beginning of the process the material was heated to the rolling temperature of ~300 °C in an induction furnace. During the rolling process the “elongation section” was executed in the first six mill stands. The speed of the first pair of rolls was 8 rpm. The finish rolling was performed in the next six stands. The diameter of the rolls was 415 mm. The oval piece was rotated by 90 ° at each stand in order to arrive in the next mill position. The bars with 1000 mm in length were continuously moved through the six mill stands. The temperature was measured by contact thermometers. The cross section of the bar was reduced from 70 mm to 34 mm during the elongation section of rolling. The rolled bars were cut to smaller pieces with 2000 mm in length. They were heated to ~300 °C before the finish rolling. In this final step of processing the diameter of the bars was reduced from 34 mm to 20 mm. The speed of the first pair of rolls in the finish rolling process was 40 rpm which is larger than in the case of the elongation section of rolling.

![Fig. 1: The shape and the size of the roll cavities](image)

The industrial manufacturing process resulted in four products. The first two were obtained by caliber rolling. The third and fourth products were obtained by the combination of caliber and flat rolling processes. In order to improve the quality of the surface and the dimensional accuracy in addition to increasing the strength of the rods, further cold rolling was performed.
Table 1: The size of the roll cavities for the twelve passes applied in industrial caliber rolling

<table>
<thead>
<tr>
<th>Cavity No</th>
<th>a (mm)</th>
<th>b (mm)</th>
<th>c (mm)</th>
<th>R1 (mm)</th>
<th>R2 (mm)</th>
<th>Cavity No</th>
<th>a (mm)</th>
<th>c (mm)</th>
<th>R1 (mm)</th>
<th>R2 (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>91.73</td>
<td>50</td>
<td>5</td>
<td>50</td>
<td>8</td>
<td>2</td>
<td>71.77</td>
<td>5</td>
<td>29</td>
<td>10</td>
</tr>
<tr>
<td>3</td>
<td>74.13</td>
<td>48</td>
<td>5</td>
<td>40</td>
<td>8</td>
<td>4</td>
<td>57.63</td>
<td>5</td>
<td>22.5</td>
<td>7</td>
</tr>
<tr>
<td>5</td>
<td>60.79</td>
<td>32</td>
<td>4</td>
<td>32</td>
<td>8</td>
<td>6</td>
<td>43.48</td>
<td>3</td>
<td>17</td>
<td>6</td>
</tr>
<tr>
<td>7</td>
<td>49.18</td>
<td>25</td>
<td>3</td>
<td>27</td>
<td>6</td>
<td>8</td>
<td>37.08</td>
<td>3</td>
<td>14</td>
<td>6</td>
</tr>
<tr>
<td>9</td>
<td>40.18</td>
<td>20</td>
<td>3</td>
<td>22</td>
<td>6</td>
<td>10</td>
<td>31.62</td>
<td>3</td>
<td>11.5</td>
<td>6</td>
</tr>
<tr>
<td>11</td>
<td>32.98</td>
<td>18</td>
<td>2</td>
<td>16</td>
<td>5</td>
<td>12</td>
<td>31.44</td>
<td>2</td>
<td>10</td>
<td>5</td>
</tr>
</tbody>
</table>

In this process the diameter was reduced by about 2 mm with a scattering of about 1 mm. The thickness of the flat bars was further reduced to 7 mm. In this process their width increased to 51 mm. The processing histories of the four industrially manufactured samples are listed in Table 2.

Table 2: The rolled industrial products

<table>
<thead>
<tr>
<th>products</th>
<th>elongation rolling</th>
<th>finish rolling</th>
<th>cold rolling</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Ø70⇒Ø34 mm/6 passes</td>
<td>Ø34⇒Ø20 mm</td>
<td>Ø20⇒Ø18 mm/2</td>
</tr>
<tr>
<td>2</td>
<td>Ø70⇒Ø34 mm/6 passes</td>
<td>Ø34⇒Ø20 mm</td>
<td>mm/6 passes</td>
</tr>
<tr>
<td>3</td>
<td>Ø70⇒Ø34 mm/6 passes</td>
<td>Ø34⇒14x45 mm</td>
<td>14x45⇒7x51 mm</td>
</tr>
<tr>
<td>4</td>
<td>Ø70⇒Ø34 mm/6 passes</td>
<td>Ø34⇒14x45 mm</td>
<td>mm/1 pass</td>
</tr>
</tbody>
</table>

3. Modelling of Rolling Process

3.1 Influence of the strain path on material flow

In the present work the objective of the modelling was to demonstrate the cyclic nature of the process, indicating that it is non-monotonic, resulting in fine grains. The permanent deformation of the samples is demonstrated by the strain trajectory approach as initiated by Ilyushin [8], representing the deviatoric strain tensor (\(\varepsilon\)) in a five-dimensional vector space as:

\[
\begin{align*}
\varepsilon_1 &= \sqrt{\frac{3}{2}} \ln V'_{11}, \\
\varepsilon_2 &= \sqrt{2} \left( \ln V'_{22} + \frac{1}{2} \ln V'_{11} \right), \\
\varepsilon_3 &= \sqrt{2} \ln V'_{12}, \\
\varepsilon_4 &= \sqrt{2} \ln V'_{23}, \\
\varepsilon_5 &= \sqrt{2} \ln V'_{31}
\end{align*}
\]

where \(\ln V'\) is the logarithmic deviatoric strain tensor. Some examples for the strain trajectories are shown in Fig.2. Trajectory #1 is monotonic, #2 is nearly monotonic, #3 is simple non-monotonic and #4 is cyclic non-monotonic.
In order to appreciate the measure of the non-monotonicity of deformation, consider the nearly monotonic trajectory #2[9]. During deformation the end-point of the strain vector travels along the curved OA trajectory. The ideally monotonic deformation corresponds to the straight line OA. Therefore, at the deformation time, $t$, the measure of non-monotonicity is given as:

$$\text{NM}(t) = \frac{\bar{\varepsilon}(t)}{\bar{\varphi}(t)} \geq 1,$$

where $\bar{\varepsilon}$ is the total equivalent strain, which is equivalent to the length of the trajectory, and $\bar{\varphi}$ is the equivalent logarithmic strain, which equals the length of the straight trajectory OA. In the case of non-monotonic deformation, the complete trajectory is separated into $n$ nearly monotonic portions. For each part the $(\text{NM})_i$ is determined as the ratio of the length of the local trajectory part and the straight segment connecting its end points. In this case the measure of non-monotonicity of the whole deformation is given as:

$$\text{NM} = \sum_{i=1}^{n} (\text{NM})_i$$

The larger the value of NM, the higher the degree of non-monotonicity of deformation. It should be noted that although all $(\text{NM})_i$ values are larger than one, NM may decrease with increasing strain as the end points of the segments may vary during the development of the deformation trajectory.

Regarding the strain path, caliber rolling has a cyclic, non-monotonic character. During the finite element modelling, the material flow was described by a constitutive equation which can consider this effect. In order to take into consideration this aspect, a measuring method and a calculation process [10,11] were developed which were used to determine the flow curves for Grade 2 titanium under cyclic, non-monotonic plastic deformation with different strain amplitudes. The schematic representation of the results is shown in Fig. 3.
Fig. 3: The schematic representation of the flow stress evolution under cyclic plastic deformation

The bold curves in Fig. 3 represent the flow curves for each individual pass of the cyclic, non-monotonic plastic deformation process, while the curves through the $\sigma_{y_{\max}}^{(i)}$ and the $\sigma_{y_{0}}^{(i)}$ values indicate the upper and the lower limiting curves of the cyclic flow stress, respectively. The extrapolation of the first flow curve ($\sigma_{y(1)}^{(1)}$) shows the equivalent stress - equivalent plastic strain relation under monotonic deformation. The cyclic equivalent curve, plotted by the dashed line, represents a monotonic flow curve which is characteristic to the total cyclic forming process. The principle of the definition was to accomplish the equality of the plastic work between the total forming process and this flow curve. Regarding the recent study, we carried out modelling experiments at constant strain rate and temperature during the deformations, but changing these physical parameters in each test. Using the resulting flow curves, the following general constitutive equation was derived to describe the cyclic flow stress ($\sigma_y$) in terms of equivalent plastic strain, equivalent plastic strain-rate, plastic strain amplitude and temperature:

$$
\sigma_y(\bar{\varepsilon}_p, \bar{\varepsilon}_p, \Delta\bar{\varepsilon}_p, T) = \sigma_{y_{r}}(\bar{\varepsilon}_p) \left[1 + C \ln \bar{\varepsilon}_p \right] \left[1 - \frac{T}{T_r} \right]^{m}
$$

where $\bar{\varepsilon}_p$ is the equivalent plastic strain, $\bar{\varepsilon}_p$ is the equivalent plastic strain-rate, $\Delta\bar{\varepsilon}_p$ is the plastic strain amplitude, $\sigma_{y_{r}}$ is the reference flow stress, and $C$ and $m$ are material constants. The normalized strain-rate and temperature in equation (4) are defined as

$$
\bar{\varepsilon}_p = \frac{\bar{\varepsilon}_p}{\varepsilon_{pr}}, \quad T_* = \frac{T}{T_r},
$$

where $\varepsilon_{pr}$ is the reference strain rate, and $T$ and $T_r$ are the current and the reference temperatures in Kelvin degrees, respectively.

### 3.2 Finite element calculation

MSC.Marc 2013.0 nonlinear finite element (FE) software was used for modelling the thermo-mechanical processes in industrial rolling procedures. Similar calculation has been done
in [12]. The rolling passes were prepared in separate models, where the initial geometrical, stress and deformation states were the results of the previous calculations. When it was necessary to reduce the calculation time, the length of the work piece was shortened, making use of the steady-state character of the process. In the model, the rolls were perfectly rigid, while the deformed material was assumed to be isotropic and elastic-plastic.

The workpiece was modelled by a three-dimensional mesh of eight-node linear brick elements. The initial mesh consisted of 18,800 elements and 21,109 nodes. Re-meshing was performed after each rolling step. The Updated Lagrange formulation was used in the FEM analysis. The constitutive equation used in FEM for the description of the dependence of flow stress on strain, strain rate and temperature was determined from cyclic multiaxial forging tests performed on Grade 2 titanium at different strain rates and temperatures.

The temperature rises due to the heat generated by plastic deformation and friction, as well as the cooling of the sample due to heat-transfer between the billet and the environment (rolls and air) were considered. The heat generated by plastic deformation was calculated as the product of the plastic work and a conversion factor of 0.9. The heat generated by friction was also calculated using the friction coefficient between the workpiece and the rolls given in terms of their relative velocities. In the calculation of the heat-transfer the rolled material was considered to be in contact with two rigid surfaces of the rolls, which have constant temperatures of 65 °C in industrial rolling.

![Fig. 4: Roll cavity filling during the caliber rolling as shown by FEM analysis](image)

The progress of the industrial caliber rolling process obtained by FEM analysis is illustrated in Fig. 4 where the roll cavity filling is shown for different rolling passes. Fig. 5 shows the total plastic strain and the parameter of non-monotonicity at five different points in the sample. The locations of these points on the cross-section of the initial rod are shown at the upper left corner in Fig. 5. Both the strain and the parameter of non-monotonicity change mainly when the rods travel through the roll cavities. The total strain increases with increasing the duration of caliber rolling in all points and it reaches a value of about 3.5-5.1 at the end of deformation. The degree of non-monotonicity first increases up to about 1.5, then it varies
between 1.2 and 2 for long rolling times. \( NM \) has a considerably higher value than one which indicates the non-monotonic nature of deformation in industrial caliber rolling.

![Fig. 5: Changing of the total strain and the parameter of non-monotonicity at points A, B, C, D and E during industrial rolling. The figures show the evolution of the parameters as a function of processing time for different numbers of passes.](image1)

In the FEM analysis the components of the logarithmic deviatoric strain tensor were determined which gave the components of the strain vector. The evolution of these components were visualised as the rolling proceeds in Fig. 6. It is revealed that considerable shear strains were developed during caliber rolling which most probably resulted in the grain refinement during deformation.

**Experimental methods**

The mechanical properties of the rolled materials were investigated by tensile tests using an Instron universal mechanical testing machine (type 8809) at room temperature and a cross-head velocity of 6 mm/min. The tests were carried out on cylindrical specimens with the length and diameter of 25 and 5 mm, respectively, machined out of the rolled bars. 3–3 samples were tested under the same conditions.

The grain structure in the initial and the rolled specimens was examined by a Tecnai G2 X-TWIN transmission electron microscope (TEM). The TEM foils were prepared from both the cross- and longitudinal sections of the rods which were thinned by mechanical grinding to a thickness of 20–40 \( \mu \text{m} \).
Fig. 6: Changing the components of the strain vector at points A, C and E during caliber rolling. The figures show the evolution of the strain vector components as a function of processing time for different numbers of passes.

The foils were further thinned by Ar-ion milling using a Gatan Model 691 precision ion polishing system. The microstructures of the initial and the rolled samples were also examined by X-ray line profile analysis. Before measurements the surface was mechanically polished to a mirror finish with diamond paste. The surface layer, distorted during polishing, was removed by chemical etching using hydrogen fluoride. The measurements of the X-ray diffraction lines were performed on the longitudinal sections using a special high-resolution diffractometer with CoKα1 radiation (wavelength: λ = 0.1789 nm). The scattered intensity was detected by imaging plates. The line profiles were evaluated using the Convolutional Multiple Whole Profile (CMWP) fitting procedure. In this method, the diffraction pattern is fitted by the sum of a background spline and the convolution of the instrumental pattern and the theoretical line profiles related to the crystallite size, dislocations and twin faults. The details of the procedure are available elsewhere [13].

5. Experimental results and discussion

The results of the mechanical tests carried out on the industrially rolled materials are shown in Fig. 7. Similar data for the materials rolled in laboratory have been published in [12]. Caliber rolling performed in laboratory resulted in a slightly higher strength and a similar ductility compared to the process carried out under industrial conditions. The reason for the slightly better mechanical performance of the material rolled in the laboratory can be attributed to a more rigorous control of the thermo-mechanical conditions (e.g. the temperature of the sample) during the manufacturing process.
Caliber rolling both in laboratory and industrial environments yielded about two times larger strength than that for the initial annealed state. The parameters characterizing the ductility do not show uniform tendencies. The elongation to failure slightly decreased due to rolling while the reduction of area remained unchanged within experimental error. At the same time, the strain energy density to fracture increased to more than twice the value characteristic of the initial state. This can be attributed to the much larger flow stress values for the rolled specimens. The various industrially manufactured samples show slightly different mechanical performances (see Fig. 7) which can be attributed to the deviations in the processing conditions.

The average crystallite sizes determined by X-ray line profile analysis are 62 and 127 nm for the laboratory and industrially rolled specimens, respectively. These values are smaller than the grain size values obtained by TEM (300-500 nm), which has already been observed for other plastically deformed metals [14]. This phenomenon can be attributed to the fact that the crystallites are equivalent to the domains in the microstructure which scatter X-rays coherently. As the coherency of X-rays breaks even if they are scattered from volumes having quite small misorientations (1-2°), the crystallite size corresponds rather to the subgrain size in severely deformed microstructures [14]. The dislocation density increased to 4.7 and 3.9 × 10^{14} m^{-2} during laboratory and industrial rolling, respectively. Although, these dislocation densities are smaller than the value obtained in titanium processed by warm ECAP [15], the grain sizes in the caliber rolled samples and the ECAP-processed specimen are similar. Therefore, it can be concluded that industrial caliber rolling is capable of the mass production of UFG titanium with UFG microstructure and high strength.

6. Conclusions

1. It was shown that warm caliber rolling carried out on Grade 2 titanium in industry yielded a UFG microstructure with high strength and good ductility.
2. Finite element modelling confirmed the cyclic non-monotonic nature of caliber rolling which is necessary for the production of a fine-grained microstructure.
3. Caliber rolling carried out in industrial environment yielded similar small grain size, high dislocation density and improved mechanical properties as the process performed in laboratory. Thus, it was proved that caliber rolling is capable of mass production of UFG Ti with high strength.

Acknowledgements

This study was supported by the Hungarian Scientific Research Fund, OTKA, Grant nos. K-100500 and 109021.

References