How severe plastic deformation at cryogenic temperature affects strength, fatigue, and impact behaviour of grade 2 titanium

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Abstract. Samples of grade 2 Ti were processed by Equal Channel Angular Pressing (ECAP), either isolated or followed by further deformation by rolling at room temperature and at 170 K. The main interest of the present work was the evaluation of the effect of cryogenic rolling on tensile strength, fatigue limit and Charpy impact absorbed energy. Results show a progressive improvement of strength and endurance limit in the following order: ECAP; ECAP followed by room temperature rolling and ECAP followed by cryogenic rolling. From the examination of the fatigued samples a ductile fracture mode was inferred in all cases; also, the sample processed by cryogenic rolling showed very small and shallow dimples and a small fracture zone, confirming the agency of strength on the fatigue behaviour. The Charpy impact energy followed a similar pattern, with the exception that ECAP produced only a small improvement over the coarse-grained material. Motives for the efficiency of cryogenic deformation by rolling are the reduced grain size and the association of strength and ductility. The production of favourable deformation textures must also be considered.

1. Introduction

Medical implants are one of the possible application of ultrafine-grained materials, and a number of reason point to Ti and its alloys as materials possessing the greatest potential. However, developments are underway to avoid the use of the ubiquitous Ti6-4 alloy due to the harmful effects of Al and V on human health. Accordingly, new Ti-based materials are now available, but complexity and cost are unfavourably high. Another solution is the replacement of the Ti6-4 alloy by the commercially pure metal, but there is a penalty in this substitution: yield strength of 350 MPa for grade 2 Ti and close to 900 MPa for the alloy. This shortcoming can be corrected by techniques of severe plastic deformation (SPD), for instance by ECAP, whose main effect is grain size refinement down to the sub-microcrystalline range [1]. By this technique the yield point of ECAP processed grade 2 Ti has been upgraded to 710 MPa, a value almost equal to that of the Ti6-4 alloy [2]. However, besides static strength the behaviour under dynamic loading conditions is critical for implant materials; unfortunately these last two properties conflict with the high strength conferred by any SPD process,
and as pointed out by Suresh good fatigue behaviour requires a favourable strength-ductility combination [3].

In the context of ECAP processed materials the knowledge of the influence of grain size on fatigue is most important; on this respect two information are relevant [4, 5]: (i) the fatigue strength of fcc metals is not affected by the grain size; (ii) but it is in materials deforming by planar slip, that is, those which do not form a cell structure. Ti deforms by planar slip and accordingly Turner and Roberts [5] have shown that its fatigue limit increases significantly with grain refinement from 100 to 9 µm. More recently, and on actual ultrafine-grained (UFG) Ti, a number of investigations confirmed the above findings [6, 7, 8]; in particular a good relationship between fatigue behavior, Hall-Petch effect and dislocation hardening was observed. Also, there is a consensus that UFG materials show only a marginal advantage over their coarse grained counterparts under cyclic stress-controlled conditions or high cycle fatigue (HCF), and even deterioration in the strain-controlled regime [9]. However, Vinogradov et al. observed that irrespective of grain size the latter conclusion does not include Ti, [10]. As for fatigue notch sensitivity, it was found to be lower for UFG Ti, a shortcoming that Semenova et al. claim that can be corrected by a post deformation heat treatment [11]. Finally, it is well known that besides static and fatigue strength, the impact properties qualify the suitability of a material for its end application. Although data on UFG Ti are still relatively scarce there are indications of a positive effect; for instance Stolyarov et al. showed that grain reduction by ECAP enhances the Charpy fracture energy of Ti, even down to low testing temperatures [12].

The present paper reports on the effect exerted by ECAP followed by cryogenic deformation upon the room temperature tensile strength, HCF and Charpy impact energy of commercially pure Ti. Results are compared with literature data and comments on the fracture mechanism are based on fracture surface observation.

2. Experimental material and procedures

An extruded bar of commercially pure Ti qualifying as grade 2 (0.09%O, 0.06%Fe, 0.001%C, 0.01%N, 0.01%H, balance-Ti) was employed. Its initial microstructure from which the grain size was estimated as equal to 15 µm is available in a previous paper [13]. After a 983 K/2 h anneal the material was machined into billets with 10 mm diameter and 70 mm length, which were inserted in a 120° ECAP die. Processing consisted of four passes at 573 K following route B_C. Half of the deformed billets were kept in the ECAP condition whilst the other half was further deformed by rolling (70% thickness reduction) at room temperature and at cryogenic temperature. For this last step the billets were immersed in liquid nitrogen from which they were removed once thermal equilibrium was established. Natural heating followed until the samples reached 170 K, which was chosen as the initial rolling temperature. Adiabatic heating has not been taken into account. ECAP processed samples were identified by the letter X preceded by a numeral indicating the number of passes; same system followed by CR or RTR identifies the samples processed by ECAP and further deformed by rolling at 170 and 300 K, respectively. Details of the tensile tests were given in ref. [13]. Smooth rectangular cross section fatigue specimens with a continuous radius between ends equal to 20 mm, having a 1.5 x 3 mm² minimum cross section and 10 mm length were spark-eroded from the processed billet and plates in such a way that their axes were parallel to the extrusion (or rolling) direction. Finally, they were mechanically polished down to R_s = 0.11 µm in order to remove any damaged layer. The fatigue tests were carried out in an INSTRON 8808 hydropulse equipment operating under sinusoidal cyclic loading at 15 Hz and R = -1, in air and at room temperature. Wöhler curves and endurance limits (σ_0) were determined for the 4X and 4XCR samples. In order to evaluate the impact energy of the UFG material, instrumented Charpy impact tests were performed at room temperature on sub-size V-notched specimens, following DIN 50115 (1 mm depth notch, 3 x 4 x 27 mm³) and employing a CEAST Charpy – IZOD impact machine with a capacity of 15 J. The specimens were extracted with their axes parallel to the corresponding ECAP billet axis and parallel to the rolling direction. The initial loading velocity was 5 m/s and data of force and hammer
displacement were continuously recorded during the tests and stored in a PC. The fracture surface of fatigued and Charpy tested specimens was observed by Scanning Electron Microscopy (SEM).

3. Experimental results

3.1. Tensile behavior

The tensile data ($\sigma_y$ = yield stress, $\sigma_u$ = ultimate stress, $\varepsilon_u$ = uniform elongation and $\varepsilon_f$ = elongation to fracture) and Vickers hardness of samples 0X (coarse-grained) 4X, 4XRTR and 4XCR were plotted against the equivalent strain imposed by the respective deformation process. Fig. 1 shows that SPD, regardless the process schedule employed produced a substantial strength enhancement, whilst ductility losses were very small.

Figure 1. Yield, ultimate strength, uniform and total elongation to fracture of grade 2 Ti in the following conditions: (i) coarse-grained – 0X; (ii) ECAP processed – 4X; (iii) ECAP processed and rolled at 300 K – 4XRTR; (iv) ECAP processed and cryorolled at 170 K – 4XCR. Process equivalent strain and Vickers hardness are indicated.

More precisely, the percentile increase of $\sigma_y$ with respect to the coarse-grained sample is 79%, 135% and 184% for samples 4X, 4XRTR and 4XCR. This is quite significant, particularly when considering that the Ti6-4 alloy exhibits yield and ultimate strength equal to 880 and 910 MPa, respectively, and 14% elongation to fracture [14]. Fig. 1 shows that the highest strength was achieved by cryogenic rolling: yield strength is 21% higher than that of the sample rolled at 300 K, whilst its total elongation is almost identical. Tensile properties higher than these here reported can be found in the literature; for instance Semenova et al. achieved $\sigma_y$ and $\sigma_u$ equal to 1100 and 1250 MPa, respectively, while maintaining a reasonable elongation [15]. However said result was obtained on grade 4 Ti and only after a number of post-ECAP processing, such as forging and drawing. It must be recalled that yield and ultimate strength of grade 4 are respectively equal to 560 and 680 MPa [14], that must be compared with the lower values of the coarse-grained grade 2 Ti, see Fig. 1.

3.2. Fatigue behaviour
Figure 2. Wöhler plots comparing fatigue life and endurance limits for grade 2 Ti in the following conditions: coarse-grained [6], processed by ECAP (4X) and by ECAP plus cryogenic rolling (4XCR). Data for the Ti6-4 alloy is included for comparison.

Fig. 2 shows that the material processed by cryogenic rolling achieved the highest endurance limit, an improvement of 30% and 98% over the ECAP processed and the coarse-grained samples. Fig. 3 gives different views of the fracture surfaces of samples 4X and 4XCR.
3.3. Charpy impact energy

Fig. 4 shows how the impact absorbed energy and the fatigue limit increase progressively with the SPD equivalent strain. SEM micrographs of the fracture surfaces are in Fig. 5, together with data on dimple size distribution. In all cases the fracture surface is typically ductile.

![Figure 3. SEM micrographs of fracture surfaces of fatigue specimens 4X and 4XCR](image)

![Figure 4. Plots of fatigue limit and instrumented Charpy absorbed energy as a function of the SPD equivalent strain for samples processed by ECAP and ECAP followed by cryogenic rolling. Results for the coarse-grained sample are included for comparison.](image)
Figure 5. SEM micrographs of representative Charpy fracture surfaces. (a) coarse-grained; (b) ECAP processed, and (c) ECAP processed and deformed by cryogenic rolling. Micrographs 1: crack initiation; micrographs 2: fracture region. The arrows indicate the shear lips width.
The instrumented Charpy test generates load-time curves from which the impact energy can be computed by integration. Fig. 6 shows the F-t curves for the 0X, 4X and 4XCR samples, and Table 1 summarizes their recorded and calculated impact energies and the geometrical parameters taken from the Charpy fractured specimens. Unfortunately, the maximum impact load of specimen 4XCR fell above the software capability and had to be estimated.

![Figure 6. Charpy test force-time curves for samples 0X, 4X and 4XCR.](image)

**Figure 6.** Charpy test force-time curves for samples 0X, 4X and 4XCR.

**Table 1.** Charpy impact energy and geometrical parameters of the fractured specimens

| Parameter / sample | 0X | 4X | 4XCR |
|--------------------|----|----|------|
| Shear lip size (mm) | -  | 0.27 | 0.77 |
| Lateral expansion (mm) | 0.24 | 0.25 | 0.30 |
| Average dimple size (µm) | 23 | 4 | 2 |
| Absorbed energy (J) | F-t curve | 2.2 | 2.6 | 4.2 |
| | machine dial | 2.5 | 2.9 | 4.2 |
| | Maximum impact force (N) | 1050 | 1110 | 2830* |

(*estimated)

It is known that from the shape of the force-times curves it is possible to infer how the total impact energy (E_t - area under the curve) partitions into crack initiation (E_i) and crack propagation (E_p). It also informs on the elastic energy dissipated in the process (E_E), the maximum force exerted by the specimen (F_max) and the energy absorbed up to the maximum force (W_max).

4. Discussion

The tensile behaviour of severely deformed Ti has been presented and discussed in ref. 13, and only the main points will be considered and expanded here. The first observation regards the positive effect of cryogenic rolling on both tensile strength and elongation to fracture. It is common sense that said properties are conflicting properties, although the literature on UFG materials shows a number of exceptions to that rule. Second, very short uniform elongation is often seen in those materials, indicating poor work hardening capacity, and the present results follow this pattern, the resistance to post-necking localized plastic flow is very high, see in Fig. 1 for the difference between \( \epsilon_i \) and \( \epsilon_{up} \) for all the samples, including the pair 4XCR and 4XRTR. Therefore, such behaviour cannot be ascribed solely to cryogenic processing. Twinning induced plasticity is capable of explaining the coexistence of strength and ductility [18, 19], but in some instances investigators failed to detect significant amounts
of said crystallographic defect in commercial purity Ti processed by ECAP [10] and followed either by room temperature [20] or by cryogenic rolling [13]. On the other hand, twinning has been observed in coarse grained [21] and in UFG Ti processed at cryogenic temperatures [22]. Finally, it is well known that hexagonal metals have a restricted number of slips systems; for instance the principal and secondary slip systems for Ti are respectively the prismatic \{1\overline{1}0\}<\overline{1}1\overline{2}0> and the basal \{0001\}<1\overline{1}0>. According to Wang and Huang, room temperature deformation by rolling activates a combination of said slip systems and produces a texture with the basal pole tiled ±20 – 40° away from the normal direction and rotated towards the transverse direction [23]. Also, the \{10\overline{1}0\} pole aligns itself with the rolling direction, while directions \{1\overline{1}2\overline{0}\} are parallel to the plate transverse direction.

In short, a definite explanation of ductility differences needs more experimental data on structural features such as texture measurements and more observation of the fine microstructure. With regard to strength, however, a reasonable explanation can be found in grain size differences, as determined in ref. 13 for samples 4X and 4XCR: 0.3 and 0.2 µm, respectively.

When fatigue tests are realized in the stress-controlled mode the results express the resistance to crack initiation, hence depend primarily on strength. Accordingly, the fatigue behaviour of SPD processed grade 2 Ti reflects the strength improvement brought about by such process, and the highest endurance limit is showed by the cryorolled sample. Moreover, in all present conditions the \(\sigma_u/\sigma_y\) ratio lies within a narrow range, viz. 0.40 - 0.46, thus confirming the role of mechanical strength on cyclic behaviour. Differences between the 4X and 4XCR fracture appearance are as follow: (i) the region of crack propagation takes up about \(\frac{1}{2}\) of the former and \(~\frac{2}{3}\) or more of the latter total surface; (ii) the dimple size is much larger in the former than in the latter sample; (iii) the average striations spacing \(S_s\) is slightly smaller in the 4X sample: 0.6 against 0.9 µm. As for observation (i) it is significant that the region of fast fracture is much smaller in the cryorolled sample, reflecting its higher strength, hence its capacity to sustain cyclic overloads. Fig. 3 shows that the region of fast fracture is typically ductile, dimples being smaller and shallower in the 4XCR sample as a consequence of its smaller grain size, thus giving substance to the role of the Hall-Petch effect on both strength and fatigue behaviour [24]. With regard to the striations spacing, which are a measure of the lengthwise crack advance per cycle, it must be recalled that the fracture surfaces here observed came from samples subjected at different stresses. It is then reasonable to affirm that higher stress mean longer cracks:

\[
\text{4X: } \sigma = 350 \text{ MPa and } S_s = 0.6 \mu\text{m}; \quad \text{4XCR: } \sigma = 640 \text{MPa and } S_s = 0.9 \mu\text{m}
\]

- and the search of any possible relationship with grain size, strength or ductility will be meaningless.

The effect of SPD on the Charpy impact strength is analogous to the exerted on fatigue behaviour. The fracture surfaces of all samples are typically ductile, with the dimple size decreasing with the process equivalent strain. In all cases, however, they are a multiple of the grain size, showing that the fracture stress must be surpassed over a critical length. From Fig. 6 and Table 1 it is apparent that the impact energy (either calculated or read from the machine dial), \(F_{\text{max}}\), and the geometrical parameters of the Charpy specimens are remarkably consistent. Again cryogenic SPD gave the best result. Examination of the force-time curves shows that: (i) data on \(E_T\), \(E_s\), \(E_P\) and \(F_{\text{max}}\) and the specimens geometrical parameters of coarse-grained and ECAP processed samples are almost identical despite the large grain size difference; (ii) for the same samples the Force - time curves shape show that crack propagation consumed most of the energy; (iii) cryogenic processing inverts the pattern, and crack initiation is more energy demanding; this is in agreement with the large shear lips observed in this sample, see Fig.5a-c and Table 1; (iv) although the \(F_{\text{max}}\) of the 4XCR sample had to be estimated, the above mentioned figure and table clearly show that it is much higher than of the two other samples, a consequence of its higher yield strength.

Unfortunately the present data cannot be quantitatively compared with the available literature due to the lack of Charpy specimen size uniformity among the various investigations.
5. Conclusions
When all other process parameters are equal, cryogenic rolling increases both strength and fatigue limit over the ECAP processed material, regardless if this is followed or not by room temperature rolling. The smaller grain size is responsible for such behaviour, but phenomena such as texturing and dislocation accumulation may play a role and should be thoroughly investigated. Instrumented Charpy tests were performed on coarse-grained, ECAP and ECAP + cryorolled samples, and again cryogenic rolling gave the highest impact value. Fatigue and Charpy broken specimens showed a ductile appearance with dimple size and depth decreasing progressively from the coarse-grained to the cryorolled sample. Analysis of the fracture surface shows that in the latter sample the region of fatigue crack propagation is larger than those of the other samples, with the initiation step consuming a large proportion of the Charpy impact energy. Both observations point out to the importance of yield strength on fatigue and impact strength.

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