LA-UR- 00-4923

Approved for public release; distribution is unlimited.

Title: Microstructure and Properties of Pure Ti Processed by ECAP and Cold Deformation

Author(s): Vladimir V. Stolyrav, Ufa, Russia
Yuntian T. Zhu, MST-STC
Terry C. Lowe, MST-DO
Ruslan Z. Valiev, Ufa, Russia

Submitted to: The 2001 TMS Spring Meeting, February 11-15, 2001, New Orleans

Los Alamos
NATIONAL LABORATORY

DISCLAIMER

DISCLAIMER

MICROSTRUCTURES AND PROPERTIES OF ULTRAFINE-GRAINED PURE Ti PROCESSED BY ECAP AND COLD DEFORMATION

Vladimir V. Stolyarov [a], Yuntian T. Zhu [b], Terry C. Lowe [b], and Ruslan Z. Valiev [a]

[a] Institute of Physics of Advanced Materials, Ufa State Aviation Technical University
Ufa 450000, K. Marksa 12, Russia
[b] Materials Science and Technology Division, Los Alamos National Laboratory
Los Alamos, NM 87545, USA

Abstract

Equal channel angular pressing (ECAP) has been used to refine the grain size of commercially pure (CP) Ti as well as other metals and alloys. CP-Ti is usually processed at about 400°C because it lacks sufficient ductility at lower temperature. The warm processing temperature limits the capability of the ECAP technique in improving the strength of CP-Ti. We have employed cold deformation following warm ECAP to further improve the strength of CP-Ti. Ti billets were first processed for 8 passes via ECAP route B_C, with a clockwise rotation of 90° between adjacent passes. The grain size obtained by ECAP alone is about 260 nm. The billets were further processed by cold deformation (cold rolling) to increase the crystalline defects such as dislocations. The strength of pure Ti was improved from 380 MPa to around 1000 MPa by the two-step process. This presentation reports microstructures, microhardness, tensile properties, and thermal stability of these Ti billets processed by a combination of ECAP and cold deformation.

1. Introduction

Equal-channel angular pressing (ECAP) has been used to process various metals and alloys, including Cu, Al and its alloys, Ni and Ti [1-12]. While Al and Cu materials have been processed at room temperature [2-11], processing commercially pure (CP) Ti at room temperature has resulted in the failure of the work piece because of the inadequate ductility of CP-Ti [13]. To improve the workability of CP-Ti, we have processed it at 400°C to 450°C with ECAP route B_C (rotation of the billet along its longitudinal axis by 90° clockwise between consecutive passes)[1]. The ECAP resulted in an average grain size about 260 to 350 nm, depending on the diameter of the Ti billet. Larger diameter leads to larger grain size, because of the slower cooling down of the Ti billet during the ECAP process [14].

ECAP significantly refined the grains and increased the strength of coarse-grained Ti by 68% [1]. To further improve the strength of ECAP-processed CP-Ti, we have combined cold deformation (cold rolling) with ECAP to further introduce crystalline defects and/or refine the grains. The cold rolling requires less workability and can process the CP-Ti at room temperature without destroying the work piece.

Recently, we used cold extrusion following warm ECAP to process CP-Ti [14]. The ECAP significantly refined the grain size and increased the yield and maximum strength to 640 MPa and 710 MPa, respectively, without changing the sample dimension. The subsequent cold extrusion further introduced higher dislocation density, and elongated the grains, which consequently increased the yield and maximum strength to 970 MPa and 1050 MPa, respectively, without ruinously changing the dimension of the Ti work piece.

Although cold extrusion has proved a feasible technique to process CP-Ti, it also has several disadvantages. These include requirement of coating lubricant before each extrusion pass, high contact friction, low surface quality, short life of extrusion die, and inhomogeneous strain in the material cross–section. As a comparison, another cold deformation technique, cold rolling, does not have the above-mentioned problems. It is more efficient and technically simpler.

In this investigation, we have combined ECAP and cold rolling to further improve the strength of CP-Ti. CP-Ti rods were first processed by warm ECAP (route B_C) to reduce the grain size to about 350 nm, and then cold-rolled at room temperature. This paper reports the processing, microstructure, mechanical properties and thermal stability of ultrafine-grained CP-Ti processed by ECAP-cold rolling (ECAP-CR).

2. Experimental Procedures

Commercially pure Ti with an average grain size of 10 μm and containing impurities including 0.12 wt% O, 0.01 wt% H, 0.04 wt% N, 0.07 wt% C, and 0.18 wt% Fe were used as the starting material. The dimensions of the starting billets are 26 mm in diameter and 120 mm long. ECAP route B_C, which rotates the work piece 90°clockwise along its longitudinal axis between two adjacent passes, was used to process the Ti billets. This route was chosen because it yields the best surface quality and more equiaxed grains than other routes[1]. The die channel angle is 90°. The entrance channel has a diameter of 26 mm and the exit channel has a diameter of 25 mm, which is slightly smaller than the entrance channel to allow easy reinsertion of the billet into the entrance channel in the following pass [14]. All billets were processed for a total of 8 passes, with the starting temperature at 450°C. The temperature dropped with each pass and reached 400°C at the 8^{th} pass. Molybdenum desulphide was used as lubricant.

The Ti billets processed by ECAP were machined into a dimension of 16 mm in diameter and 80 mm long to fit the dimension of the rollers and to have a smooth surface finish. The billets were then rolled and its cross-section was changed from round to oval to square. The rolling speed is 0.1 m/second. Each rolling pass decreased the dimension in the normal direction for 0.5 to 1 mm. Self-heating during the rolling heated the Ti billet to about 100°C. Therefore, the billet was cooled in cold water to room temperature after each rolling pass. The total rolling strain, calculated as the total reduction in cross-section area, was either 35% or 55%.

Transmission electron microscopy (TEM) samples were cut from both the transverse-section and the longitudinal-section. These samples were prepared by jet electropolishing. Bright and dark field TEM images were taken using a JEM-100B microscope. An accelerating voltage of 100 KV was used. Electron diffraction patterns were taken from an area of 2 μm^2.

The microhardness was measured in both transverse and longitudinal sections by the Vicker's method under a load of 100 g for 10 seconds. Ten measurements were made for each sample and their average was taken as the microhardness of the sample.

Tensile tests were performed at room temperature using a universal testing machine IR-5047-50. To study the thermal stability, some Ti billets processed by ECAP followed by cold rolling for 35% strain were annealed at 200°C to 400°C for half an hour. Cylindrical samples [1] with a gauge section of 5 mm in diameter and 25 mm long were machined from the as-processed and annealed Ti billets. A displacement rate of 1 mm/min. was used for all tests. Yield strength, ultimate strength, elongation to failure, and reduction in area at the necking cross-section were measured.

3. Results

3.1 Microstructures

Figures 1 shows the TEM micrographs and selected area diffraction (SAD) pattern of Ti billet processed by ECAP route B_C for 8 passes. Figure 1a was taken from the transverse section of the as-processed billet while Fig. 1b was from the longitudinal section. The SAD pattern, which was taken from the transverse section, indicates the existence of large fraction of high-angle grain boundaries. Clustered diffraction spots suggests the existence of low–angle grain boundaries. Such a mixture of low-angle and high-angle grain boundaries is typical of ECAP-processed metals and alloys [2-4, 15-19]. The average grain size as measured from the transverse section (Fig. 1a) is

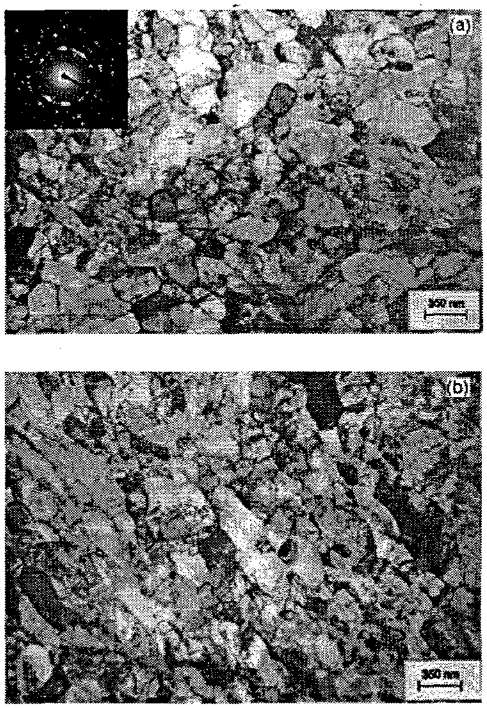

Figure 1: TEM micrographs from (a) transverse section and (b) longitudinal section of a Ti billet processed by ECAP routes B_C for 8 passes. The initial diameter of the billet was 26 mm.

about 280 nm. It can be seen from Fig. 1b that the grains are somewhat elongated in the longitudinal direction.

Figures 2 and 3 show the microstructures of Ti billets processed by ECAP followed by cold rolling at room temperature for a rolling strain of 35% and 55%, respectively. No intermediate annealing was performed during the cold rolling. Figure 2a, which is from the transverse section, show an average grain size of 170 nm, which is smaller than the grain size after ECAP (280 nm). Comparing Fig. 2a with Fig. 1 revealed that cold rolling introduced much more dislocations into the CP-Ti work piece. In addition, the grains are becoming less equiaxed in the cross-section after cold rolling (Fig. 2a). Figures 2b shows that the grains are elongated in the longitudinal section. High-density dislocations and dislocation networks are shown inside grains. It appears from Fig. 2 that 35% cold rolling only elongated the equiaxed-grains produced by ECAP without little further grain refinement.

Further cold rolling to a strain of 55% made the grains less equiaxed and more irregular in the cross-section, as evidenced by elongated and angular shapes of some grains (Fig. 3a). In addition, the grain boundaries are less sharp, and in many cases very blurred. For these reasons, it is hard to measure the grain size from the cross-section. But the average grain size measured from the cross-section should be smaller in CP-Ti billet subjected to 55% rolling strain than those subjected to 35% rolling strain, because of further grain elongation. Figure 3b shows that the grains are further elongated after 55% rolling than after 35% rolling strain (Fig. 2b). In addition, strong fragmentation is clearly shown in the elongated grains. These

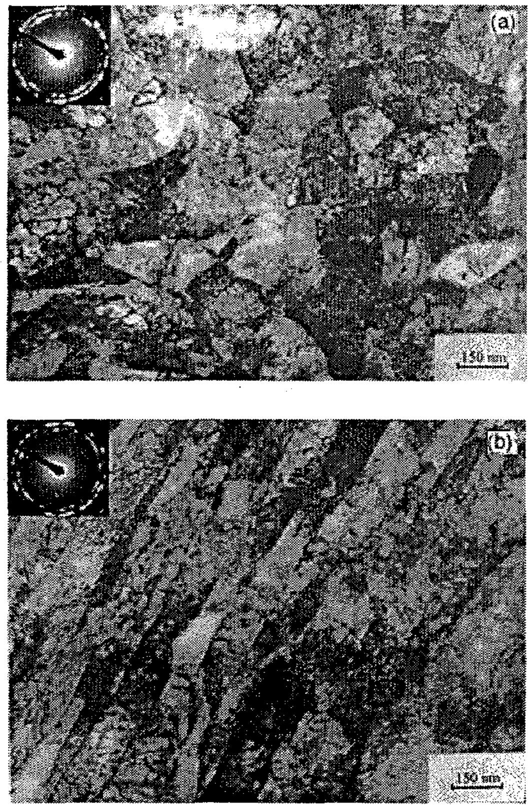

Figure 2: TEM micrographs and SAD patterns of a Ti billet processed by ECAP route B_C for 8 passes, followed by cold rolling for a strain of 35%. (a) transverse section and (b) longitudinal section.

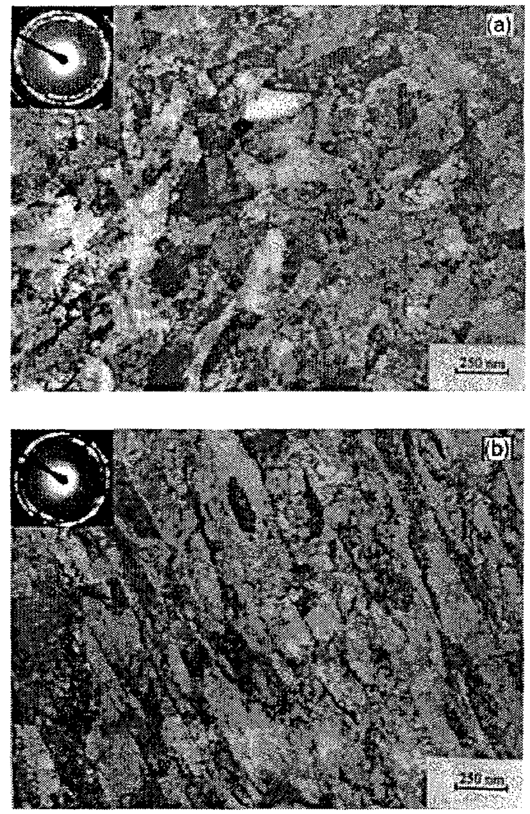

Figure 3: TEM micrographs and SAD patterns of a Ti billet processed by ECAP route B_C for 8 passes, followed by cold rolling for a strain of 55%. (a) transverse section and (b) longitudinal section.

fragments are likely to have low angle misorientation with each other. The azimuth spreading of the diffraction spots is about 5° to 7°, which indicates the existence of high internal stress. Bright field TEM micrographs from both the cross-section (Fig. 3a) and the longitudinal section (Fig. 3b) show high dislocation densities.

3.2 Mechanical Properties

3.2.1 Microhardness

Listed in Table 1 are the microhardnesses of the Ti billets under various processing states. Each microhardness value is an average of at least 10 measurements. The scatter of the measured value is about 5% or less. It is obvious that cold rolling after the ECAP has further increased the microhardness. Also, the microhardness increases with increasing strain from the cold rolling. Not surprisingly, cold rolling alone yielded lower microhardness than not only ECAP + cold rolling, but also ECAP alone. Another observation is that the microhardness is anisotropic, with the longitudinal section having higher microhardness than the transverse section for all deformation states.

Table 1. The microhardness measured from both transverse and longitudinal sections of Ti billets in various processing states.

Deformation state	Microhardness (MPa) in	
	Transverse Section	Longitudinal section
ECAP*	2700	2810
Cold rolling (35% strain)	2610	2650
ECAP + Cold rolling (35% strain)	2830	2970
ECAP + Cold rolling (55% strain)	2950	3250

* Route B_C for 8 passes

3.2.2 Tensile properties

Tensile tests were conducted on cylindrical samples with gauge dimensions of 5 mm in diameter and 25 mm long. Readers are referred to [1] for more details on the specimen geometry. The longitudinal axis of the sample coincide with that of the billets. Three samples were tested for each processing state, with at least 2 successful tests. The average mechanical properties are listed in Table 2, which shows that

ECAP increased the yield and maximum strength to 640 MPa and 710 MPa, respectively. After a cold rolling for 35% strain, the yield and maximum strength were increased to 940 MPa and 1040 MPa, respectively. Further cold rolling to a strain of 55% yielded even higher the yield and ultimate strength. As a comparison, cold rolling alone for 35% strain yielded strengths comparable to those of samples processed by ECAP.

Table 2. Yield strength ($\sigma_{0.2}$), ultimate strength (σ_u), elongation to failure (δ), and reduction in area (ψ) of Ti in various processing states. Each data is an average of 2 to 3 tests.

Processing state	$\sigma_{0.2}$ (MPa)	σ_u (MPa)	δ (%)	ψ (%)
Coarse-grained (grain size = 10 μm)	380	460	27	69
ECAP (8 passes, route B_C)	640	710	14	61
Cold rolling (35% strain)	660	670	16	48
ECAP+Cold rolling (35% strain)	940	1040	6.8	45
ECAP+Cold rolling (55% strain)	1020	1050	6	30

3.3 Thermal stability

To study the thermal stability, Ti billets processed by ECAP followed by cold rolling for a strain of 35% were annealed at 200°C to 500°C for half an hour. Cylindrical samples with a gauge dimension of 5 mm in diameter and 25 mm in length were tested under tensile mode at a displacement rate of 1 mm/minute. Shown in Fig. 4 are yield strength, ultimate strength and elongation to failure with increasing temperature. As shown, there is no significant drop in strength at annealing temperatures up to 300°C. On the other hand, annealing at 300°C for half an hour doubled the elongation to failure from 6% to 12%. More increase in elongation to failure and significant drop in the yield and ultimate strength are observed when the as-processed CP-Ti billet was annealed at 400°C for half an hour. Therefore, the strengths of the CP-Ti processed by ECAP + cold rolling is thermally stable at temperatures up to 300 °C, which is similar to the strength of CP-Ti processed by ECAP + cold extrusion, but 100°C lower than the strengths of CP-Ti processed by warm ECAP alone.

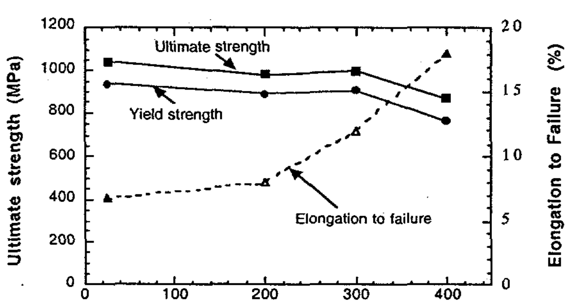

Figure 4: Thermal stability of a Ti billet processed by ECAP route B_C for 8 passes, followed by cold rolling for a strain of 35%. The annealing duration is half an hour.

Figure 5 shows the TEM micrographs of CP-Ti annealed at 400°C for half an hour following the ECAP+ cold rolling (35% strain). Figure 5a is from the transverse section. As compared with the as-processed state (Fig. 2a), the annealing changed the grains from irregular shape to equiaxed shape, and significantly reduced dislocation density. Extinction contour, which does not exist in the as-processed state, appears near many grain boundaries, attesting the relief of internal elastic strain. No significant grain growth is observed. More significant microstructural changes are observed in the TEM images from the longitudinal section (Fig. 5b). The fibrous grains in the as-processed state (Fig. 2b) became almost equiaxed after the annealing at 400°C (Fig. 5b). Dislocation density, although lower than in the as-processed state (Fig. 2b), is still very high in the annealed state. This suggests that the cells and subgrains with low-angle misorientations were formed during the

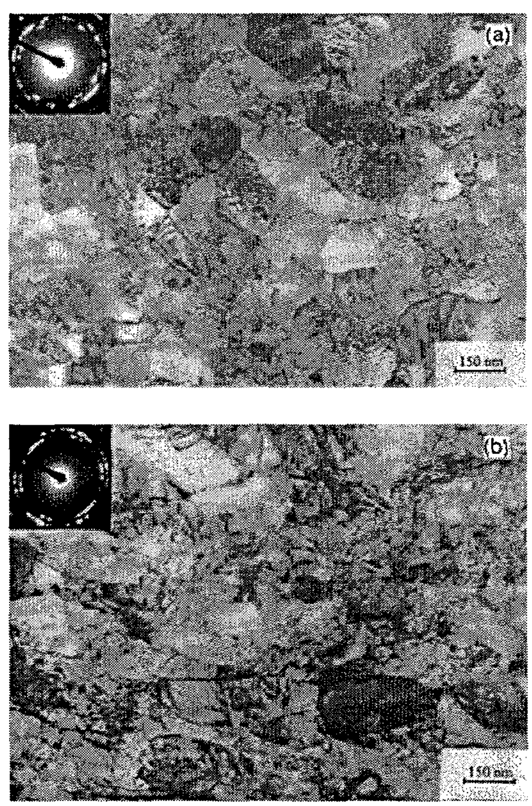

Fig. 5 TEM micrographs and SAD patterns of a Ti billet processed by ECAP route B_C for 8 passes and cold rolling for a strain of 35%, and then annealed at 400°C for half an hour. (a) transverse section and (b) longitudinal section.

annealing [18]. This is also consistent with the selected area diffraction (SAD) pattern (Fig. 5b), which shows clustered diffraction spots.

4. Discussion

This study has developed the combination ECAP and cold rolling as a new technique that can process CP-Ti into an ultrafine-grained state with high strength. This technique is superior to ECAP + cold extrusion, reported in our previous study [14], since it does not require lubrication before each rolling pass and yields better surface quality.

Due to the relatively high processing temperature (450°C-400°C), ECAP has limited capability in refining grains and improving the strength of CP-Ti. On the other hand, cold deformation such as cold extrusion and cold rolling cannot retain the geometry of the work piece, resulting in the over-reduction of one or more dimensions of work piece. In our previous work [14], a combination ECAP + cold extrusion was used successfully to process CP-Ti, and strength higher than Ti-6Al-4V alloy was obtained. However, even with the application of lubricant coating before each extrusion pass, cracks were still introduced to the surface of some work pieces. In addition, a significant extrusion strain, which equals the reduction in cross-section area, is required to obtain the high strength. It is desirable to use smaller reduction in cross-section area to obtain higher increase in strength. Figure 6 compares the yield and ultimate strengths of CP-Ti with different cold rolling and extrusion strains. It is clear that cold rolling is more effective than cold extrusion in improving the strength. It requires less cold rolling strain to reach a desired strength level. In other words, for the same strength requirement, less reduction in the cross-section area is needed by cold rolling than by cold extrusion.

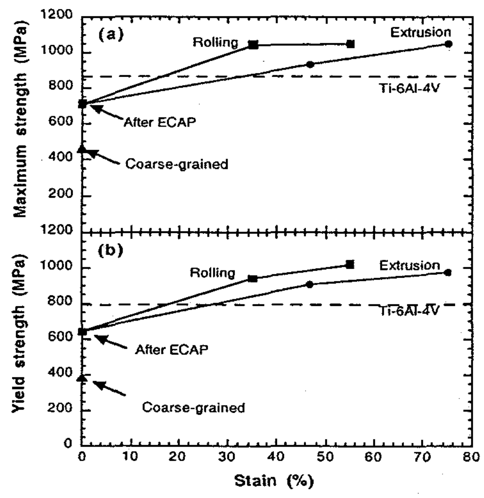

Figure 6: Comparison of yield strength and ultimate strength of Ti billets processed with varying cold rolling and extrusion strains.

Cold rolling for a strain of 35% after the warm ECAP significantly increased the dislocation density and resulted in elongated grains (Fig. 2). The grains as viewed from the cross-section become smaller and irregular in shape (Fig. 2a). Increasing strain to 55% further made the grain shape more irregular and elongated (Fig. 3). Strong fragmentation was formed in the elongated grains, indicating the formation of subgrain cells (Fig. 3b). Grain boundaries become more blurred after the CP-Ti was subject to 55% of rolling strain. No extinction contours near grain boundaries was observed in the cold rolled samples, indicating the existence of high internal elastic strains.

The strength of the CP-Ti processed by ECAP + cold rolling (35% rolling strain) is stable at temperatures up to 300 °C (Fig. 4), which is similar to CP-Ti processed by ECAP + cold extrusion. Thermal annealing at 300°C improved the ductility (elongation to failure) from 6% to 12% without significantly decreasing the strength. Annealing at 400°C resulted in a 16% decrease in ultimate strength and 19% decrease in the yield strength (Fig. 4). TEM micrographs show that in the transverse section grain shape changed from irregular to equiaxed without much change in the average grain size (Fig. 5a). High-density dislocation can still be seen in most grains, indicating the little if any recrystallization. This suggests that grain boundary migration, instead of large-scale recrystallization, changed the grain shape to more equiaxed shape. As shown in the TEM micrographs in the longitudinal section (Fig. 5b), cell structures with equiaxed shape and low-angle misorientations are formed, and the fibrous structure in the as-processed state (Fig. 2b) becomes less distinct after the annealing. These microstructural changes suggest that the strength drop after annealing at 400°C was caused mostly by the recovery process.

5. Summary

We have combined ECAP and cold rolling to process ultrafine-grained CP-Ti, and obtained yield and ultimate strengths as high as 1020 MPa and 1050 MPa, respectively. Annealing at 300°C improved the elongation to failure by 100% with only very little decrease in strength. Cold rolling does not require lubricant coating and yields better surface quality than cold extrusion. In addition, it is also more effective in improving the strength of CP-Ti than cold extrusion, requiring less cross-sectional reduction to obtain the same strength level. TEM micrographs show that cold rolling introduced higher dislocation density, and elongated the grains, and deformed grains from equiaxed shape to irregular shape as viewed from the transverse section of the Ti billet. This study proved the combination of ECAP + cold rolling as a better technique than ECAP + cold extrusion for producing ultrafine-grained materials with high strength for structural applications.

Reference

1. V.V. Stolyarov, Y.T. Zhu, I.V. Alexandrov, T.C. Lowe, and R.Z. Valiev, "Influence of ECAP routes on the Microstructure and Properties of Pure Ti," Mater. Sci. Eng. A (in press).

2. S. Ferrasse, V.M. Segal, K.T. Hartwig, and R.E Goforth, Metall. Mater. Trans. 28A (1997) 1047.

3. S. Ferrasse, V.M. Segal, K.T. Hartwig, and R.E. Goforth, J. Mater. Res., 12 (1997) 1253.

4. P.B. Prangnell, A. Gholinia, and V.M. Markushev, "Overview and Outlook for Materials Processed by Severe Plastic Deformation," Investigations and Applications of Severe Plastic Deformation, Edited by T.C. Lowe and R.Z. Valiev, Kluwer Academia Pub., Dordrecht, 2000, pp. 65-71.

5. Y. Iwahashi, Z. Horita, M. Nemoto and T.G. Langdon, Acta Mater., 46 (1998) 3317.

6. K. Oh-Ishi, Z. Horita, M. Furukawa, M. Nemoto and T.G. Langdon, Metall. Mater. Trans. 29A (1998) 2011.

7. K. Nakashima, Z. Horita, M. Nemoto and T.G. Langdon, Acta Mater., 46 (1998) 1589.

8. Y. Iwahashi, Z. Horita, M. Nemoto and T.G. Langdon, Metall. Mater. Trans. 29A (1998) 2503

9. T.G. Langdon, M. Furukawa, Z. Horita, and M. Nemoto, JOM, 50 (June) (1998) 41.

10. Y. Iwahashi, M. Furukawa, Z. Horita, M. Nemoto and T.G. Langdon, Metall. Mater. Trans. 29A (1998) 2245.

11. Y. Iwahashi, Z. Horita, M. Nemoto and T.G. Langdon, Acta Mater., 45 (1997) 4733.

12. V.M. Segal, Mater. Sci. Eng., A197 (1995) 157.

13. S.L. Semiatin, V.M. Segal, R.E. Goforth, N.D. Frey, and D.P. Delo, Metall. Mater. Trans. 30A (1999) 1425.

14. V.V. Stolyarov, Y.T. Zhu, I.V. Alexandrov, T.C. Lowe, and R.Z. Valiev, "Microstructure and properties of pure Ti processed by ECAP and cold extrusion," Mater. Sci. Eng. A (in press).

15. F.J. Humphreys, P.B. Prangnell, J.R. Bowen, A. Gholinia and C. Harris, Phil. Trans. R. Soc. Lond. A, 357 (1999) 1663.

16. B.E. Warren, X-ray Diffraction, Dover, New York, 1990.

17 I.V. Aleksandrov and R.Z. Valiev, The Physics of Metals and Metallography, 77 (1994) 623.

18 F.J. Humphreys and M. Hatherly, Recrystallization and Related Annealing Phenomena, Pergamon, Oxford, UK, 1995, p. 127-128.

19 G.W. Nieman, J.R. Weertman, and R.W. Siegel, J. Mater. Res., 6 (1991) 1012.