Explaining absence of texture development in cold rolled two-phase Zr–2.5 wt% Nb alloy

M. Kiran Kumar ^a, I. Samajdar ^{a,*}, N. Venkatramani ^b, G.K. Dey ^c, R. Tewari ^c,
D. Srivastava ^c, S. Banerjee ^c

^a Department of Metallurgical Engineering and Materials Science, Indian Institute of Technology Bombay, Powai, Mumbai-400076, India

^b ACRE, Indian Institute of Technology Bombay, Powai, Mumbai-400076, India

^c Materials Science Division, BARC, Trombay, Mumbai-400085, India

Abstract

In the present study, two distinct starting microstructures of Zr–2.5 wt% Nb have been used: (1) single-phase α hcp martensitic structure; and (2) two-phase, 10% bcc β and rest hcp α , Widmanstätten structure. In the second case, two types of α were present—near grain boundary predominantly single-phase α (about 5% of the total α) and α plates in an apparently continuous β matrix. Both (1) and (2) had similar starting crystallographic texture of the hcp α phase and were deformed by unidirectional and cross rolling. In the two-phase structure the changes in the bulk texture on cold rolling was found to be insignificant, while in the single-phase material noticeable textural changes were observed. Taylor type deformation texture models predicted textural changes in single-phase structure but failed to predict the observed lack of textural development in the two-phase material. Microtexture observations showed that α plates remained approximately single crystalline after cold rolling, while the β matrix underwent significant orientational changes. Relative hardening, estimated by X-ray peak broadening, was observed mainly in β phase; while aspect ratio of α plates remained unchanged with cold rolling—indicating absence of effective macroscopic strain in the hcp α plates. Based on microstructural and microtextural observations, a simple model is proposed in which the plastic flow is mainly confined to the β matrix within which the α plates are subjected to ‘in-plane rigid body rotation’. The model explains the observed lack of textural developments in the two-phase structure.

Keywords: Zirconium; Texture; Microtexture; Cold deformation; Texture modeling; Microstructure; Two-phase

1. Introduction

Cold deformation in crystalline material is expected to have two apparent effects on texture

development—macroscopic rotation of individual grains and development of deformation texture [1–3] and microscopic in-grain rotations or microtexture developments and corresponding substructural changes [4–9]. The latter may be accommodated by plastic instability [8], creation of new grain boundaries, lattice curvatures [9] and dislocation

substructure [4,6,7], ultimately accounting for an overall microstructural development [6,7]. In single-phase cubic systems, study of deformation texture development and quantitative texture predictions are almost routine [2,3], though an understanding of microtexture developments remains far from complete [4–9]. Two-phase commercial cubic systems usually deal with non-shearable second phase and shearable primary phase [10–14]. Relative randomization of soft primary phase around the hard second phase has been explained [10], though precise experimental verification seems pending [11].

Lower symmetry systems are more complex. In hexagonal crystals, deformation behavior strongly depends on the c/a ratio [15]. Development of deformation texture naturally depends on the effective macroscopic strain and strain tensor, available slip–twin systems and their respective critical resolved shear stresses (CRSS) [16]. Among the hcp materials, Zr alloys are widely used in structural applications in nuclear industry [17,18]. Zr–2.5 wt% Nb, in particular, is extensively used as pressure tubes in Pressurized Heavy Water Reactors (PHWR) [19]. Zirconium alloys have, in general, less than ideal c/a ratio and exhibit a strong texture development during fabrication [20]. The crystallographic texture and the microstructure usually play major roles in determining in-reactor performance of Zr alloys [17,18,21–30].

Zr–2.5 wt% Nb is usually fabricated by hot extrusion followed by either cold drawing or two steps of cold pilgering with an intermediate annealing [17,31,32]. Studies on texture developments during cold working of Zr–2.5% Nb are mainly limited to the determination of two-dimensional pole figures and not in terms of orientation distribution functions (ODFs) [20,28,33–38]. Microtexture development in cold deformed single phase and two-phase Zr alloys, has largely remained as an uncharted territory.

The classical Taylor type models of deformation texture developments [1,2,33,34,39–45,53] typically considers single-phase systems, notable exception being modeling the so-called texture randomization in two-phase commercial alloys of cubic systems [10]. The usual minority phase in Zr–2.5Nb is the bcc β . Though the difference in

flow stresses of the α and the β phase is less pronounced than in commercial cubic systems, β can be considered as shearable and α as semi-shearable [19,51,52]. The role, if any, of the shearable minority phase (β) on the deformation texture development of the majority phase (α) and if such a role can be ‘captured’ by Taylor type deformation simulations are not really known [33,40–50]. The broad objectives of the present study were to identify and understand such issues through characterization of macrotexture and microtexture of deformed Zr–2.5Nb in the single (α) and the two-phase ($\alpha + \beta$) microstructural states.

2. Experimental methods

2.1. Material and processing

The composition of the Zr–2.5 wt% Nb alloy used in the present study is shown in Table 1. Two distinct microstructures [31,54,55] were obtained from the following heat treatments:

1. Solutionizing in the β phase at 1000 °C for 30 min followed by water quenching to produce single-phase α hcp martensitic structure (a treatment often denoted as β -quenching).
2. Solutionizing in the β phase followed by annealing in $\alpha + \beta$ at 600 °C for 1/2 h to produce a two-phase Widmanstätten structure.

These were cold rolled by unidirectional and cross rolling, where cross rolling involved equal strains in alternating rolling and transverse directions. Reductions of 40, 60 and 80% were obtained in a laboratory rolling mill for the two-phase structure. For the single-phase structure a maximum reduction of about 50% could be obtained.

Table 1
Chemical composition of Zr–2.5% Nb alloy as used in the present study

Nibobium (wt%)	Oxygen (ppm)	Iron (ppm)	Hydrogen (ppm)	Nitrogen (ppm)
2.51	1092	1250	<10	30

2.2. Bulk texture measurements and deformation texture simulations

Bulk texture measurement data were obtained by measuring four incomplete pole figures of the following poles: $\{0001\}$, $\{10\bar{1}0\}$, $\{10\bar{1}2\}$, and $\{11\bar{2}0\}$. These were selected specifically to avoid, to the best possible extent, overlapping of any neighboring X-ray peaks or poles coming from either the hcp or the bcc phases. Direct measurement of the basal pole was avoided, as $\{0002\}$ is too close to $\{01\bar{1}1\}$. Measurements were made for the background as well, using usual conventions [39,53,56]. However, given the proximity of poles and presence of two-phases in both bulk and powder samples, it was felt that background values are often on the higher side and may contain X-ray counts from neighboring peaks.¹ Thus correcting raw pole figures with background may remove some of the actual texture components. Results presented in this study are hence background ‘non-corrected’. It is to be noted, however, that the overall *trend* in bulk texture developments are *similar* for background corrected and non-corrected data. X-ray ODFs were calculated by inversion of the four incomplete pole figures and using the standard series expansion [39,53,56,57].

Texture of undeformed material, in each case, was discretized to about 3000 individual orientations [58] and these were then used for subsequent deformation texture simulations. Table 2 shows the slip and twinning systems and the relative CRSS values as used by different researchers [33,41,42,44] in predicting the cold worked texture developments in Zr-based alloys. These were broadly classified as four modes, see Table 2, and were used in subsequent texture simulations. Experimental and simulated ODFs were compared visually and by respective fiber intensities or randomness numbers [39].

¹ Cold rolling also involved strong peak broadening in hcp α , which on the other hand may also bias the background measurements.

2.3. Microscopic and XRD peak-broadening characterizations

Optical microscopy samples were mechanically polished and chemically etched using a solution of 5% hydrofluoric acid and 45% nitric acid, rest being distilled water. Lateral force microscopy (LFM) samples were electropolished at -15°C and 20 V dc, while for Transmission electron microscopy (TEM) samples electropolishing, -30°C was used. The same electrolyte (30 parts perchloric acid, 170 parts *n*-butyl alcohol, and 300 parts methanol) was used in both cases.

Microstructural studies, by optical and LFM, were carried out in all three cross-sections—rolling plane (containing rolling and transverse direction or RD and TD), short transverse (ST, with ND (normal direction) and TD) and long transverse (LT, with RD and ND). LFM studies were obtained using a Thermomicroscope Autoprobe CP-R. A Philips CM200 TEM, 200 keV operating voltage, was used for microtextural studies. Microtexture measurements were obtained online using the commercial TSL-ACT package, developed by TSL-EDAX. Size measurements were obtained using online interface of CM200 series, which uses deflector coil movements to measure distances in the microstructures. At least four samples were used in each condition for obtaining microtexture information in TEM.

A direct measurement of β hardness is difficult, given the small dimensions of β and requirements of extensive etching which may bias microhardness measurements. Peak broadening of α and β were estimated, using the broadening at half-maximum intensity and the usual parabolic fit [59]. These were estimated for $(10\bar{1}2)$ α peak and (200) β peak, in order to establish differences, if any, in relative hardening behavior of the two phases [59].

3. Results

3.1. Starting microstructures and textures

Optical microstructures of single-phase and the two-phase Zr–2.5%Nb alloy are shown in Fig. 1(a) and (b). As shown in Fig. 1(a), the β -quenched

Table 2

Slip-twin combinations and their relative CRSS values, as used by different researchers for deformation texture simulations in Zr alloys

Mode	Slip-twin combinations	CRSS ratio for slip-twin combinations	Reference
A	P + TT + CT	0.8:1:2	[41]
B	P + (PY + PY1 + PY2) + TT	1:2.25:2.13	[42]
C	B + P + PY1 + PY2 + TT + CT	0.5:0.1:0.8:1.5:1:2	[44]
D	B + P(PY + PY1 + PY2) + TT	0.2:0.1:1.2	[33]

The conventions used for slip and twinning systems are: Basal (B) $(0001)\langle 1\bar{2}10 \rangle$, Prism (P) $(10\bar{1}0)\langle 1\bar{2}10 \rangle$, Pyramidal $\langle a \rangle$ (PY) $(10\bar{1}1)\langle 1\bar{2}10 \rangle$, Pyramidal $(c + a)/a$ (PY1) $(10\bar{1}1)\langle 2\bar{1}\bar{1}3 \rangle$, Pyramidal $(c + a)/b$ (PY2) $(2\bar{1}\bar{1}2)\langle 24\bar{2}3 \rangle$, Compressive Twin (CT) $(21\bar{1}2)\langle 21\bar{1}3 \rangle$ and Tensile Twin (TT) $(10\bar{1}2)\langle 10\bar{1}1 \rangle$.

alloy had a typical martensitic structure [18,19,31] containing the hcp α phase. The two-phase structure clearly had Widmanstätten morphology, see Fig. 1(b), in which packets of Nb-depleted α plates are distributed in a continuous β matrix of Nb content of about 15 wt%. The grain boundary region contained negligible volume fraction of β and is referred as only α containing region. All three cross-sections show an apparent chaotic arrangement of α plates; in other words, they are not aligned in any specific direction. Chemical etching has the problem [19,31] of etching the β region quite deep, which makes the β volume fraction appear larger than actual. This may bias microstructural observations both in optical and in scanning electron microscope (SEM). TEM may provide an alternative [31], but on restricted sample areas. In the present study atomic force microscope (AFM) in the LFM mode was used as a more accurate and quantitative tool for microstructural characterization on large electropolished samples. LFM mode was observed to be most attractive in characterizing the two-phase regions, as compared to other AFM modes. The LFM image of the undeformed two-phase alloy is shown in Fig. 1(c). As shown in the figure and verified by several LFM scans, the two-phase starting microstructure contained about 5% of only α containing region and about 10% of bcc β . As observed by TEM based microtexture measurements (see Fig. 2), individual α plates were nearly of a single orientation. The β

matrix² in-between the α plates were also almost single crystalline, though misorientations of about 1.5° on average, not exceeding $3\text{--}4^\circ$, were observed in both β matrix and α plates. In different regions (inside the same pre-transformation β grain), β matrix had ‘slightly’ different orientations, as in Fig. 2(c), which might have been caused by transformation induced strains on the retained β .

As in Figs. 3(a) and 4(a), the starting bulk textures, represented by respective ODFs, of hcp α , were similar in both single and two-phase alloy. The starting textures can be generalized or simplified as fibers—orientations with a particular crystallographic plane but all possible directions. An orientation in Euler space has unique or several Euler angles based on symmetry [39]. A fiber is a series of orientations generalized to a single class for identification—the origin of such a fiber may have links with prior processing [2,6,10,24,39]. In the present case, however, drawing such a link is premature at this stage, the fiber generalization was mainly for convenience. Some of the generalized fibers are marked in Fig. 3(a) for subsequent comparison. Both single and two-phase alloys were rolled by unidirectional and cross rolling and

² Though β is the minority phase; its apparent continuity and more importantly the fact that parts of pre-transformation β grain(s) got converted into α plates while rest remained as β ; led to the present generalization of β as the matrix phase.

Fig. 1. Starting optical microstructures: (a) single-phase β -quenched martensitic structure and (b) two-phase Widmanstätten structure of bcc β phase (black) in hcp α (white) in all three cross-sections (rolling plane, long transverse and short transverse). In (b) RD, TD and ND, as used in subsequent cold rolling, are shown. (c) LFM (lateral force microscope) image of undeformed two-phase structure, showing presence of near grain boundary only α containing region and grain interior $\alpha + \beta$ region.

developments of bulk textures of hcp α , deformation microstructure and microtexture were investigated.

3.2. Deformation texture developments in hcp α

3.2.1. Experimental results

Fig. 3 shows the bulk texture developments in two-phase microstructure, while Fig. 4 shows the same in single phase. As shown in Fig. 3 for two-phase, the individual fibers remained after extensive unidirectional as well as cross rolling. There

Fig. 2. Microtexture of undeformed two-phase $\alpha + \beta$ region. (a) Micrograph of hcp α and bcc β (appearing as the dark phase), (b), and (c) are the respective inverse pole figures representing the local orientation measurements in α and β s (for the regions of β matrix marked as i and ii in Fig. 2(a)) corresponding to (a). Undeformed α or neighboring β matrix, as in (a), appeared as nearly single orientation(s). In (a) RD and TD, as used in subsequent cold rolling, are shown.

were only slight modifications in such fibers—relative intensities at specific fiber locations (or at particular crystallographic directions) changed but overall fiber appearance remained. This point can be further elaborated by Fig. 5, representing changes in the basal fiber or $(0002)\langle uvw \rangle$ as a function of cold rolling. Arrows are used against individual orientations to indicate relative increase or decrease as compared to the undeformed material.

In single-phase structure, strong textural changes were observed (see Fig. 4). The typical fiber texture of the undeformed state did change completely, as many of the fibers or orientations vanished and/or evolved towards new orientations. Experimental observations (ODF plots) can be summarized as: insignificant textural changes in the two-phase structure, but a strong texture development in the single-phase material.

3.2.2. Deformation texture simulations

As shown in Fig. 6, simulations always predicted strong texture changes in both single and

Fig. 3. Cold deformation textures of two-phase structure. X-ray ODFs (constant ϕ_2 sections) of (a) undeformed alloy, (b) (a) after 80% unidirectional rolling and (c) (a) after 80% cross rolling. Following fibers are marked on (a): Basal $(0002)\langle uvw \rangle$ ($\phi_1 = 0-90^\circ, \phi = 0^\circ$ and $\phi_2 = 0^\circ$), Prismatic $(10\bar{1}0)\langle uvw \rangle$, Pyramidal $(11\bar{2}0)\langle uvw \rangle$, $(01\bar{1}2)\langle uvw \rangle$, $(\bar{2}4\bar{2}1)\langle uvw \rangle$ and $(15\bar{6}5)\langle uvw \rangle$. Following contour levels (for randomness levels) were used for ODF plotting—0.8, 1.0, 1.2, 1.6, 2.0, 2.5, 3.0, 4.0, 5.0 and 6.5.

two-phase³ alloy. Combinations of slip–twin and CRSS (as in Table 2) or constraints [2,53] could *not* simulate absence of texture changes, or the retention of the original fiber texture (as in Fig. 3), typical of two-phase Zr–2.5 Nb, as shown in Fig. 6(a). Strong texture changes in single phase, however, could be predicted, as shown in Figs. 4(b)

and 6(b). Fig. 4(b) is actual ODF (for single phase), while Fig. 6(b) is simulated. They are *not* exactly identical. These observations form the basis of the main inference coming out of the deformation texture simulations—combinations of slip–twin systems alone cannot explain the retention of fiber texture or the absence of texture changes in two-phase material. Such an inference is not based on the accuracy of prediction of deformation texture.

³ As pointed out earlier in Section 1 Taylor type simulations are typical of single-phase systems and one of the objectives of the present study was to test their applicability in the present two-phase structure.

Fig. 4. Cold deformation textures of single-phase structure. X-ray ODFs of (a) undeformed alloy, (b) (a) after 50% unidirectional rolling and (c) (a) after 50% cross rolling. ODF conventions are same as in Fig. 3.

Fig. 5. Change in relative strength or $f(g)$ (randomness number) of Basal fiber $(0002)\langle uvw \rangle$ in two-phase Zr alloy with (a) unidirectional and (b) cross rolling. Fiber intensity is plotted at different ϕ_1 .

3.3. Deformed microstructures in two-phase

Microstructural developments in two-phase are outlined in Figs. 7 and 8. As shown in Fig. 7(a), cold deformation by unidirectional rolling had two apparent effects: (1) The α plates align along the RD on the rolling plane. (2) Grain boundaries, which contain predominantly α plates, maintained continuity. The apparent chaotic structure in the other two cross-sections (LT and ST) remained, that is the α plates did not realign in any specific direction.

At the latter stages of deformation (60% or higher), microcracks were observed only on such

Fig. 6. Simulated ODFs for: (a) two-phase material after 80% unidirectional rolling, using full constraint Taylor model with slip–twin combination A (as explained in Table 2). (b) Single phase material after 50% unidirectional rolling, using full constraint Taylor model with slip–twin combination B (see Table 2). ODF conventions are same as in Fig. 3.

α containing region(s). Cross rolling did not align the α plates on the rolling plane and often discontinuity in grain boundary α regions was observed (see Fig. 7(b)). Perhaps it is interesting to note that apparent ductility in cross rolling was significantly better, as visible cracking was observed at 60% reduction in unidirectional rolling but not even at 80% reduction in cross rolling.

To appreciate the possible effects of rolling on the softer β matrix, LFM measurements were undertaken. Fig. 8(a) and (b) show LFM images of $\alpha + \beta$ containing regions in undeformed state and after 60% unidirectional rolling. As shown in the figures, cold rolling had ‘thinned’ β considerably though dimensions of α plates did not change.

Fig. 7. Two-phase structure (a) in all three cross-sections after 60% unidirectional rolling and (b) on the rolling plane (TD being the vertical direction) after 60% cross rolling. In (a) RD, TD and ND are shown. Alignment of α plates on rolling plane and along RD was observed, while no such alignment was apparent at other two cross-sections.

This was observed only on the rolling plane⁴—on average from about 300 to about 90 nm drop in thickness of β . Fig. 8 clearly shows an inhomogeneous deformation with softer β matrix clearly

⁴ Typically during rolling with strain homogenization, one may expect an increase or a decrease in respective dimensions along rolling and normal directions. However on the rolling plane, containing rolling and transverse directions, a decrease in dimension(s) is not expected.

Fig. 8. LFM (lateral force microscope) images of $\alpha + \beta$ containing region in (a) undeformed and in (b) 60% unidirectionally rolled material. Structures were obtained from rolling plane, RD being indicated in (b) by the apparent alignment of α and β phases.

accommodating more plastic deformation. Similar observations on ‘thinning’ of β matrix was made in TEM as well (see Figs. 2(a) and 9(a)).

3.4. Microtextural developments

The purpose of the microtextural studies was to obtain a broad understanding of the deformation in individual regions, such as only α containing and $\alpha + \beta$, and to use such understanding in explaining observed differences in macrotextural developments between single and two-phase structures. The microtextural developments were generalized as two parameters:

- Point to point misorientation development—as estimated from point to point orientation measurements in α and in β (i.e. measuring misorientation between adjacent points).
- Long range misorientation (LRM) [60] development.

As in a previous study [60], the LRM development was measured by considering lines along RD inside individual α plate and adjoining β matrix and then estimating gradual changes in misorientation along these lines with respect to the starting point or origin. Such an LRM can be generalized as cumulative or repetitive [60]. In the first case

overall orientation is expected to change or evolve, while the second case indicates a lack of orientation change or an orientation stability [60]. Observation on $\alpha + \beta$ and only α containing regions are presented separately in Figs. 9 and 10.

In $\alpha + \beta$ containing regions, strong orientation changes were observed in the β matrix and insignificant orientation changes in the α plate (see Fig. 9(b) and (c)). Respective misorientations in individual α plate and in adjoining β matrix were about $5\text{--}8^\circ$ and above 15° (often exceeding 20°). As in Fig. 9(d) and (e), LRM development in a single α plate was almost always repetitive, while in adjoining β matrix cumulative LRM was often observed. Based on limited TEM statistics, it appears that the β matrix got randomized through cold work. This point, though interesting, is not linked to the main theme of the present study, textural changes in hcp α , and hence is not discussed further.

In only α containing region (or in single-phase structure), stronger misorientation developments were observed and LRM developments were often cumulative, see Fig. 10. Large misorientation developments (exceeding 20°) in predominantly α containing regions were always across some deformation heterogeneities, as shown in Fig. 10. Such trends in misorientation and LRM, as in Figs. 9 and 10, were observed at all reductions and both in unidirectional and cross rolling.

Fig. 9. Microtexture in $\alpha + \beta$ region after 80% unidirectional rolling. (a) Bright field micrograph, rolling (RD) and transverse (TD) directions being marked; (b) and (c) are the respective inverse pole figures showing the local orientation measurements in an α plate and adjoining β matrix; (d) and (e) show the LRM development on the marked lines in (a), respectively for α and β . For LRM, individual local orientation measurements were obtained along RD at an approximate spacing of about 300 nm apart, using spot size of 100 nm or less, total length of such orientation measurements were at least 3–4 μm (and often more).

Fig. 10. Microtexture in only α containing region after 80% unidirectional rolling. (a) Micrograph showing α region with deformation heterogeneities. (b) Inverse pole figure representing the local orientation measurements in (a). (c) LRM development on the marked line in (a), conventions being same as in Fig. 2.

3.5. Relative deformation in α and β

3.5.1. Relative hardening of α and β

As shown in Table 3, peak broadening (with increase in reduction) of α and β had marked difference. Noticeable peak broadening in β and insignificant broadening in α was observed. The peak broadening is expected to scale with relative hardness [59] and hence can be considered as an index of hardening. The results indicate stronger hardening in β , though reaching saturation at 40% reduction, and insignificant hardening in α (both being estimated macroscopically). It is to be noted that the effects were similar in other α and β peaks as well (this was confirmed by measurements at extreme reductions).

Table 3
Relative peak broadening in α and β phase

Sample two-phase Zr-2.5 wt% Nb (%)	Peak width (in deg on 2θ) (α -(10 $\bar{1}$ 2))	Peak width (in deg on 2θ) (β -(200))
0	0.5	0.8
40	0.7	3.1
60	0.6	3.2
80	0.5	3.32

Measurements were obtained for (10 $\bar{1}$ 2) α and (200) β peaks at 0.01°/s scan rate. Broadening of peaks were estimated at half maximum intensity using standard parabolic fit [59]. Respective deformation percentages (by unidirectional rolling) is given in the table. It is to be noted that, in single phase, α had typically shown noticeable broadening of (10 $\bar{1}$ 2) peak with increase in strain.

3.5.2. Effective macroscopic strain in α plates

The dimensions of α plates did not change with reduction. This was confirmed by all the microscopic techniques used in the present study. In the starting two-phase structure, and as viewed on the rolling plane (and also on LT plane, though with relatively limited statistics), the α plates had an average aspect ratio (on the rolling plane) of 5.33:1 (average length and thickness being 8 and 1.5 μm , respectively). *No noticeable* change in this average aspect ratio of α was observed with increasing reduction. This, on the other hand, strongly indicates absence of effective macroscopic strain in α plates.

4. Discussion

4.1. Expanding on the experimental observations

The main experimental observations need to be summarized before any effort of explanation(s) is made:

1. Bulk texture of hcp α —insignificant changes in two-phase, but strong developments in single phase. The former seems to be unexplainable by any combination (as in Table 2) of slip–twin or CRSS. Microtexture—in $\alpha + \beta$ containing region, β appeared to be deforming or

developing new orientation(s), while individual α plate(s) remained essentially single crystalline.

2. Relative orientation of α plates—in unidirectional rolling, there was an overall alignment of α plates along RD on the rolling plane, while no such alignment was observed, by optical microscopy, on other two cross-sections (LT and ST).
3. Relative deformation of α and β —peak broadening: insignificant hardening in α but noticeable hardening in β (though reaching saturation at 40% reduction). As the dimensions and the aspect ratio of α plates remained unchanged, the plates did not undergo any effective macroscopic strain.

In a word, results strongly indicate absence of significant deformation (or deformation leading to an effective macroscopic strain) in α plates, only possibilities of ‘alignment’ along rolling direction (and only on rolling plane) did exist. Hence reorientation or texture development, if any, is expected to originate from such alignment and not from slip and twin. Next two paragraphs are aimed at expanding and explaining these results further.

Absence of texture change during deformation may not indicate failure of deformation texture modeling, but a relative orientation stability [2]. However, the starting orientations or texture of hcp α was clearly unstable; this has been illustrated quite well by the textural changes in single phase and also by texture simulations in two-phase. Apparent stability of the two-phase evidently had something to do with the softer β phase. It should be pointed out that qualitative observation on the lack of texture development in cold worked two-phase Zr–2.5 wt% Nb has also been reported [33], though possible explanation(s) was absent.

Microstructural observations in two-phase clearly indicate two distinct regions—minority (about 5%) only the α containing region and the majority $\alpha + \beta$ region. It is apparent that textural changes in the latter will be decisive. In the $\alpha + \beta$ region, α plates were ‘floating’ on a softer β matrix. The individual plates were single crystalline (at least on the level of TEM based local orientation measurements). Deformation was largely

restricted to β matrix—after deformation individual α plates remained single-crystalline and their average dimension (or aspect ratio) did not change, they only got ‘aligned’ along the RD and on the rolling plane, see the schematics of Fig. 11(a) and also Figs. 7 and 9. Such an ‘alignment’ does not require slip and twin, but only a ‘rigid body rotation’, as shown schematically in Fig. 11(a).

The low energy α/β boundaries, which are formed at the stage of α precipitation from β matrix, has a strong tendency to be retained in spite of heavy deformation of the matrix [19,31]. In order to accommodate the heavy deformation it is, therefore, essential for the α plates to undergo rigid body rotation in a soft continuous β matrix, without causing any effective macroscopic strain to the

Fig. 11. The in-plane rigid body rotation model: (a) schematic diagram illustrating rotation(s) on the rolling plane and along RD, of α plates in a continuous β matrix. An imaginary unit cell represented few of the α plates, both before and after deformation. It is to be noted that the shades shown in these unit cells are just to give a sense of necessary rotation and does not correspond to the indicated crystallographic planes. The schematic overestimates β fraction, but otherwise dimension tolerances were kept in consideration for 60% reduction. (b) The simulated, with random in-plane rigid body rotation, ODF of the two-phase structure. Absence of texture development in (b) corresponds to the experimental observations in two-phase structure. (c) Simulated ODF for 20% deformation (by unidirectional rolling) simultaneously with in-plane rigid body rotation or random ND rotation. Random ND rotations were applied after small incremental strain (2%). The ODF fails to capture lack of texture development typical of deformed α plates in two-phase material.

plates. The physical driving force for the proposed deformation mode is derived from the difference in interfacial energy of α/α and α/β boundaries. Insignificant changes in the line broadening of α peaks and no change in the dimensions or aspect ratio of α plates, clearly support such a mechanism.

4.2. On the possible mechanisms accounting for deformation texture of α plates

There are three possible mechanisms to account for the developments of bulk deformation texture in α .

- Deformation through slip and twin: the observed lack of change in the aspect ratio of α (with reduction) signified absence of *effective* macroscopic strain in α plates. The misorientation developments did indicate straining of α , perhaps because of inter-plate interactions. Such strains were, however, local or microscopic and could have cancelled each other. In regions of strong inter-plate interactions, local strains did create changes in misorientation and orientations—effects canceling each other in absence of an effective macroscopic strain and hence did not contribute to the bulk texture developments. Such an argument seems logical, and does explain the repetitive local orientation developments in microscopically strained α plates, along with no net change in bulk texture. An insignificant role of slip–twin systems (in deformation texture development), due to an absence of effective macroscopic strain, also justify the results of Taylor type deformation texture simulations.
- Deformation through rigid body rotation: ND rotation of the α plates were observed, see Fig. 7(a). As discussed subsequently ‘in-plane rigid body rotation’ or simple ND rotation does explain retention of the α fiber texture.
- Combination of both deformation and rigid body rotation: a combination of deformation and in-plane rigid body rotation is also possible. This has been discussed in the subsequent section.

4.3. Model for deformation texture of α plates

Taylor type deformation texture simulations are gross statistical estimation of the rotation of individual crystallites, assuming or imposing strain homogeneity [1–3,53,61]. Though such models are incapable of predicting possible rotation or orientation development in an individual grain [61], but are reasonably successful in predicting the general reorientation of a large number of crystallites [1–3,53,61], even in hexagonal system [40–45]. In general, Taylor models do not take any microstructural feature in account and are valid for single phase. The notable exception being application of upper bound analysis with Taylor type approach [10,62] in modeling the texture development of soft matrix phase around hard and non-shearable second phase. The situation is approximately reverse here—grossly the idea is to predict or explain texture development in hard second phase.

As generalized by the simple schematics of Fig. 11(a), deformation only involved ‘alignment’ of α plates along the RD and on the rolling plane—an in-plane rigid body rotation. This is supported by the microstructural observations (see Fig. 7(a)) that the α plates did not align in any specific directions on the other two cross-sections, LT and ST, and there were no dimensional changes in α plates, as established by no changes in aspect ratios in any of the cross-sections (especially the rolling plane). Such in-plane rigid body rotation can be simplified as rotation around ND or a rotation on ϕ_1 (Euler angle in Bunge notation). This rotation cannot change the α plane(s) parallel to the rolling plane, but can alter the crystallographic directions. To test such a model, random in-plane rigid body rotation(s) were simulated on the starting texture (or 3000 discretized orientations). This was attempted by imposing random ϕ_1 rotations between 0 and 90°, the range of angular realignment necessary for α plates. As shown in Fig. 11(b), deformation(s) involving in-plane rigid body rotation cannot eliminate individual fibers which represent orientations with same crystallographic plane but all possible crystallographic directions. However, ϕ_1 rotation will alter intensities along a given fiber. Such a model seems to explain the

experimental observations of the two-phase structure.

A simultaneous combination of in-plane rigid body rotation (or random ND rotation) and deformation could not explain the lack of texture development in α . Small strain increment of 2% followed by random ND rotations also brought noticeable changes in simulated texture at above 6% reduction. Such a simulated ODF for 20% deformation by unidirectional rolling is included in Fig. 11(c). The ODF shows significant texture changes, contrary to the experimental observations (as explained in Section 3.2.1.). In a word, *only* in-plane rigid body rotation seems capable of explaining absence of texture developments in α plates.

The simulated ODF (imposing in-plane rigid body rotation on α) as well as microstructural observations on all three cross-sections have clearly shown that the α plates did rotate around the ND during cold rolling. Fig. 7(a) has not shown any alignment of the α plates along RD in ST and LT planes. This observation is in variance with what is observed in many cases of two-phase materials. In this context, it is worth mentioning that the present Zr–Nb alloy differs from other two-phase materials in many ways. For instance, in this alloy the minority β phase is the matrix phase and macroscopic deformation is primarily confined to this phase. The α plates, on the other hand, mainly undergo in-plane rigid body rotation and no effective macroscopic strain.

Since there is no compressive deformation along any direction on the rolling plane, rotation of α plates around ND will not cause expulsion of the matrix β phase from the intervening gaps between the α plates. On the other hand, on ST and LT planes, rotation of α plates will necessarily squeeze out the β phase present between the α plates, causing formation of α/α interfaces at the expense of α/β interfaces. As has been reported earlier [31], the formation of α/β boundaries in this alloy is energetically favorable in comparison with α/α interfaces. The β phase always acts as the continuous wetting medium in which the α plates are embedded and deformation processes always tend to maintain the β phase between the α plates. Such a deformation process is consistent with the α plate

rotation only around ND while rotation around RD and TD is not favorable.

The other interesting issue is texture development in the ‘matrix’ β . The development of β texture was not amenable for a detailed study because of its small volume fraction, high strain and finely divided distribution. The only method of texture determination in deformed β is TEM based local orientation measurements. The results of such measurements are presented in Fig. 9(c), which shows the relative randomization of the β (compare Figs. 9(c) and 2(c)). This technique, however, suffers from inherently limited statistics. The randomization of β may be explained from near random α plate interactions.

The apparent thinning of β in the rolling plane, as observed experimentally (see Fig. 8), may also be explained from realignment process of α plates, as shown schematically in Fig. 11(a). In the random distribution of α plates, often the β thickness (between two α plates) may appear larger than in the aligned α structure (as expected or observed after rolling). The schematic (see Fig. 11(a)), though it overestimates β fraction, tries to bring out this point.

It needs to be pointed out that two-phase Zr–2.5 wt% Nb is formed by cold pilgering—a tube forming operation involving reduction of tube wall and of tube diameter. A ratio of these two reductions is called the ‘Q-factor’ [63]. According to the present model, while the former reduction involves in-plane rotation, the latter involves out-of-plane rotations (rotations on ϕ and ϕ_2) and hence may strongly affect crystallographic texture. General observation and understanding in nuclear industry is large Q-factor does not change texture but small Q-factor does—an observation qualitatively explainable from the present concept or model.

5. Summary

Starting structures: In the present study, two distinct starting microstructures were used—single-phase (hcp α') and two-phase (hcp α and bcc β) structure. The latter contained about 10% bcc β phase, the structure being divided in two distinct

regions—only α containing (about 5% by area) and $\alpha + \beta$ Widmanstätten structure. A gross definition of the Widmanstätten structure is single crystals of hcp α plates in an apparently continuous β matrix. The hcp α in both cases had similar starting texture. Both structures were deformed by cold rolling to different reductions. Following are the brief summary on the textural, microstructural and microtextural changes, along with an attempt to explain and to relate such changes.

1. Texture development in hcp α : Development of the deformation texture was noticeable in single-phase alloy, but insignificant in two-phase. Taylor type deformation texture models, with different combinations of slip–twin systems (including CRSS) and constraints, could not predict observed absence of texture changes (or retention of original α fiber texture) in two-phase Zr–2.5 Nb alloy.
2. Microstructural and microtextural developments: The majority of the two-phase structure consists of Widmanstätten α in a β matrix. Unidirectional rolling aligned the α plates along rolling direction and only on rolling plane, but individual α plates remained nearly single crystalline.
3. Relative deformation in α and β : α Plate dimension (or the aspect ratio) did not change with deformation, signifying absence of effective macroscopic strain in α . Relative hardening in β , as observed by X-ray peak broadening, was significant, while insignificant hardening or peak broadening was observed in α .
4. Explaining the absence of bulk texture developments in two-phase: Three possible mechanisms of bulk deformation texture in hcp α can be expected:
 - Deformation through slip and twin—absence of effective macroscopic strain in α plates does eliminate dominant role of slip–twin in deformation texture developments. This, on the other hand, explains inability of Taylor type deformation texture models in explaining bulk texture developments of two-phase material.
 - In-plane rigid body rotation—such an effect was observed by microscopy. Such a mech-

anism or model, though simplistic, explains absence of quantitative texture development in rolled two-phase alloy.

- Combination of in-plane rigid body rotation and deformation—a combination failed to explain absence of quantitative texture development.

Acknowledgements

The authors will like to acknowledge financial support from Board of Research in Nuclear Sciences (BRNS) in conducting this research and Department of Science and Technology (DST) support in acquiring the TSL-ACT package. The authors will like to acknowledge NFC (Nuclear Fuel Complex, Hyderabad, India) for providing the Zr–2.5 wt% Nb alloy used in the present study. The authors will also like to acknowledge use of Regional Sophisticated Instrumentation Centre (RSIC), IIT Bombay, TEM facility. The X-ray texture facility in IIT Bombay is a gift from KU Leuven, Belgium. Indradev Samajdar will like to express his gratitude to KU Leuven and to Prof. P. Van Houtte.

References

- [1] Taylor GI. *Inst. J Inst Metals* 1938;62:307.
- [2] Aernoudt E, Van Houtte P, Leffers T, Plastic deformation and fracture of materials. In: Mughrabi H, editors, *Materials science and technology: a comprehensive treatment*. Cahn RW, Haasen P, Kramer EJ, editors. vol. 6. VCH: Weinheim, Germany; 1993. p. 89.
- [3] Vanoutte P, Delannay L, Samajdar I. *Tex Micro* 1999;31:109.
- [4] Bay B, Hansen N, Hughes DA, Kuhlmann-Wilsdorf D. *Acta Metall Mater* 1992;40(2):205.
- [5] Hughes DA, Hansen N. *Acta mater* 1997;45(9):387.
- [6] Nandedkar VM, Samajdar I, Narashiman K. *ISIJ Int* 2001;41(12):1517.
- [7] Kuhlmann-Wilsdorf D. *Acta mater* 1999;47(6):1697.
- [8] Dillamore IL, Roberts JG, Bush AC. *Metal Sci* 1979;13:73.
- [9] Winther G, Juul Jensen D, Hansen N. *Acta mater* 1997;45(12):5059.
- [10] Van Houtte P. *Acta Metall Mater* 1995;43(7):2859.
- [11] Samajdar I, Rabet L, Verlinden B, Van Houtte P. *ISIJ Int* 1998;38(6):539.

- [12] Ashby MF. *Philos Mag* 1970;21:399.
- [13] Humphreys FJ. *Acta Metall* 1977;25:1323.
- [14] Engler O. *Mater Sci Technol* 1996;12:859.
- [15] Numakura H, Koiwa M. *Metall Sci Technol* 1998;16:4.
- [16] Akhtar A. Ab. *J Nucl Mater* 1973;47:79.
- [17] Charles OS. *Nuclear reactor materials*. In: Reading, MA: Addison-Wesley; 1967. p. 130.
- [18] Maussner G, Ortlieb E, Weidinger H-G. *Materials for nuclear reactor core applications*. In: London: BNES; 1987. p. 49.
- [19] Srivastava D, Dey GK, Banerjee S. Technical Report. Bhabha Atomic Research Centre, Bombay; 1992. BARC/1992/I/011.
- [20] Linga Murty K, Ravi J, Mahmood ST. *Nucl Eng Des* 1994;148:1.
- [21] Prat F, Grange M, Besson J, Andrieu E. *Metall Mater Trans* 1998;29A:1643.
- [22] Bai JB, Ji N, Gilbon D, Lebrun JL. *Scr Metall Mater* 1992;26:369.
- [23] Lee DH, Kwun SI. *Scr Metall Mater* 1994;31(11):1475.
- [24] Hwang SK, Ryoo HS, Morris Jr. JW. *Metall Trans* 1991;22A:2247.
- [25] Holt RA. *J Nucl Mater* 1979;82:419.
- [26] Ibrahim EF, Holt RA. *J Nucl Mater* 1980;91:311.
- [27] Holt RA, Ibrahim EF. *Acta Metall* 1979;27:1319.
- [28] Cheadle BA, Eells CE, Evans W. *J Nucl Mater* 1967;23:199.
- [29] Ibrahim EF. *J Nucl Mater* 1983;118:260.
- [30] Perovic V, Weatherly GC, Simpson CJ. *Acta Metall* 1983;31(9):1381.
- [31] Srivastava D, Dey GK, Banerjee S. *Metall Mater Trans* 1995;26A:2707.
- [32] Holt RA, Aldridge SA. *J Nucl Mater* 1985;135:246.
- [33] Salinas Rodriguez A, Jonas JJ. *Metall Trans* 1992;23A:271.
- [34] Salinas-Rodriguez A. *Acta Metall Mater* 1995;43(2):485.
- [35] Tenckhoff E. *Metall Trans* 1978;9A:1401.
- [36] Bechade JL, Bacroix B, Guillen R. *Icotom* 10. Bunge HJ, editor. *Mater Sci Forum* 1994;10(157-162):617.
- [37] Le Roux SD, Van Der Merwe DJ. *Icotom* 10. Bunge HJ, editor. *Mater Sci Forum* 1994;10(157-162):1455.
- [38] Brandão LPM, da Costa Viana CS. *Icotom* 10. Bunge HJ, editor. *Mater Sci Forum* 1994;10(157-162):1365.
- [39] Bunge J. *Texture analysis in materials science*. London: Butterworths, 1982.
- [40] Lebensohn RA, Sanchez PV, Pochettino AA. *Scr Metall Mater* 1994;30:481.
- [41] Lebensohn RA, Tomé CN. *Acta Metall Mater* 1993;41(9):2611.
- [42] Turner PA, Tomé CN. *Acta Metall Mater* 1994;42(12):4143.
- [43] Tomé CN, Lebensohn RA, Kocks UF. *Acta Metall Mater* 1991;39(11):2667.
- [44] Fundenberger JJ, Philippe MJ, Wanger F, Esling C. *Acta mater* 1997;45(10):4041.
- [45] Thornburg DR, Piehler HR. *Metall Trans* 1975;6A:1511.
- [46] Chirkin AV, Al-Nakow AS. *J Nucl Mater* 1991;183:62.
- [47] Pochettino AA, Gannio N, Vial Edwards C, Penelle R. *Scr Metall Mater* 1992;27:1859.
- [48] Isayenkova MG, Perlovich YuA. *Phys Met Metall* 1991;71(5):81.
- [49] Song SG, Gray III GT. *Acta Metall Mater* 1995;43(6):2325.
- [50] Hutchinson JW. *Metall Trans* 1977;8A:1465.
- [51] Banerjee S, Dey GK, Srivastava D, Ranganathan S. *Metall Mater Trans* 1997;28A:2201.
- [52] Luo CP, Weatherly GC. *Metall Trans* 1988;19A:1153.
- [53] Van Houtte P. *Tex Micro* 1988;8-9:313.
- [54] Perovic V, Weatherly GC. *Acta Metall* 1989;37(3):813.
- [55] Perovic V, Weatherly GC, Fleck RG. *Can Metall Quart* 1985;24(3):253.
- [56] Toth LS, Van Houtte P, Fundenberger, JJ, Wanger F. *Manual of MTM-KUL (hexagonal ODF software)*, MTM-KULeuven, editor. Belgium; 1992.
- [57] Van Houtte P. *Manual of MTM-FHM*, MTM-KULeuven editor. Belgium; 1995.
- [58] Toth LS, Van Houtte P. *Tex Micro* 1992;19:229.
- [59] Cullity BD. *Elements of X-ray diffraction*. In: 2nd ed. Reading, MA: Addison-Wesley; 1978. p. 460.
- [60] Samajdar I, Ratchev P, Verlinden B, Aernoudt E. *Acta mater* 2001;49:1759.
- [61] Samajdar I, Verlinden B, Van Houtte P. *ISI Int* 1998;38(7):759.
- [62] Rabet L. PhD Thesis, ISBN No: 90-5682-021-4, Katholieke Universiteit Leuven; 1996 [in Dutch].
- [63] Jeffrey CB, Linga Murty K. *Symposium on Zirconium Alloys for Reactor Component (ZARC-91)*. BARC, Library and Information Services, BARC, Mumbai, India 1991, 1.