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Deformation and Fracture of Alpha Zirconium Alloys

(The Ductility of Zirconium and Its Alloys)

by

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DEFORMATION AND FRACTURE OF ALPHA ZIRCONIUM ALLOYS

by

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J.A. Jensen* and W.A. Backofen**

ABSTRACT

The deformation and fracture of Zircaloy-4 and pure zirconium sheets having strong (0002) textures were studied under plane-strain compression at temperatures from 20° to 800°C. Zircaloy-4 underwent a ductility transition from limited strain (0.1 to 0.3) to extensive fracture-free flow (> 0.6) at about 300°C; pure zirconium was ductile at all temperatures. These differences were traced to others in deformation mechanisms. Below 300°C, Zircaloy-4 accommodated compression along [0001] by $\langle c + a \rangle$ slip; these dislocations were mainly confined to thin bands oriented close to $\{11\bar{2}4\}$. In pure zirconium $\{11\bar{2}2\}$ twinning followed by second and third-order retwinning tended to distribute strain more uniformly. Above 300°C both materials deformed by a combination of $\{10\bar{1}1\}$ twinning and $\langle c + a \rangle$ slip.

Fracturing in Zircaloy-4 was by highly localized shear along thin bands of $\{11\bar{2}4\}$ orientation. From an analysis based on measured strain-hardening behavior and standard heat flow equations, it is suggested that localized shear can be brought about by thermal softening during slip-band formation.

⁺ This paper is based on a thesis submitted to Massachusetts Institute of Technology by J.A. Jensen in partial fulfillment of the requirements for a Ph.D. degree.

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DEFORMATION AND FRACTURE OF ALPHA ZIRCONIUM ALLOYS

INTRODUCTION

Plasticity in hexagonal close packed metals depends on the operation of a number of different slip and twin systems. Differences in the stresses for activating these systems, together with the large lattice rotations associated with twinning, cause textures to form early in any reduction history. In sheet material the texture tends to be (0002). Rolling then becomes a process of forcing plane-strain reduction along the c-axis. The occasional need for doing this is met in all HCP metals, and the most obvious ways of coping with it are by twinning and slip on systems with $\langle c + a \rangle$ Burgers vectors. How successfully that is done (success being fracture-free reduction) varies considerably from material to material. Experience with Zircaloy has shown that fracturing can be a problem under such conditions of loading, which contrasts with the extensive flow of pure zirconium identically loaded and with large reductions-in-area of Zircaloy pulled in tension in the plane of the sheet (1-3).

The means by which Zircaloy or, for that matter, pure zirconium accommodates reduction along the c-axis are not clear, even though much is known about their deformation mechanisms. In Zircaloy, tensile width-to-thickness strain ratios coupled with texture measurements indicate that prism slip, $\{10\bar{1}0\} \cdot \langle 11\bar{2}0 \rangle$, is active (4-6); no other slip systems

have been identified. Pure zirconium also glides on prism planes (7,8), and evidence for basal (9) and pyramidal slip (8) has been reported as well. None of these systems, however, have a $\langle c \rangle$ component in their Burgers vectors. Twinning has been observed in both Zircaloy (10,11) and pure zirconium (7,8), although the twin planes have been identified only in the latter. Twinning on $\{10\bar{1}2\}$ and $\{11\bar{2}1\}$ leads to extension along the c-axis while $\{11\bar{2}2\}$ twinning has the opposite effect (7); the $\{11\bar{2}3\}$ twinning shear has never been determined, but twinning on these planes is too infrequent to be very important (7). Of the reported deformation mechanisms then, only $\{11\bar{2}2\}$ twinning will support a c-axis reduction, and it has only been observed in pure zirconium at temperatures below 300°C (7).

From the record it is uncertain how either Zircaloy or pure zirconium deforms during processing. Both materials, however, are often shaped by rolling so that some structural means for reduction along the c-axis must be available. Whether or not the lower ductility in Zircaloy is a reflection of deformation mechanisms uniquely its own is not at all clear. Accordingly, it was decided to study the deformation and fracture behavior of these materials under conditions that would simulate processing, and these were chosen to be plane-strain compression at temperatures from 20° to 800°C.

EXPERIMENTAL PROCEDURES

Materials

One material was a commercially produced Zircaloy-4 (coded hereafter as Zr-4) received as annealed 3/32-in. thick plates; the other was pure zirconium (I-Zr) prepared from iodide crystal bars. Chemical analyses for each are given in Table 1. The general fracturing behavior of Zr-4 was determined with material having a 15 to 25 μm grain size (measured as a mean linear intercept). This small grain size, while typical of Zr-4, prevented quantitative measurements of deformation and fracture features. Therefore some material was coarsened by a critical-strain and anneal technique (2.9% strain followed by 50-hr at 810°C) which produced grains up to 2-mm in diameter while maintaining a uniform dispersion of precipitates. The grains in I-Zr were coarsened by a similar technique.

Basal-plane pole figures for each material appear in Fig. 1. Texture in the fine-grained Zr-4 was determined by the Schulz reflection method (12) with $\text{Cr K}\alpha$ radiation. Contours with 1 $\equiv$ randomness were drawn by normalizing intensities after the method of Grewen et al. (13). The grain size in the coarsened Zr-4 and I-Zr would not permit standard X-ray techniques. A two surface analysis (14) based on polarized light measurements (15) was used to orient the basal planes of 100 grains in each of these materials.

TABLE 1
CHEMICAL ANALYSES, BY WEIGHT*

Element	Zr-4	I-Zr
Sn	1.36%	350 ppm
Fe	0.17%	30
Cr	0.083%	20
Al	< 1000 ppm	50
Cu	< 100	3
Mg	< 100	100
Mn	< 100	40
Mo	< 1000	trace
Ni	< 10	10
Pb	< 10	---
Si	< 100	500
Ti	< 100	80
Hf	----	70
O	1200	80 to 120
N	8	5
H	10	6
C	230	28

* Analysis for metallic impurities in I-Zr was furnished by the supplier, Foote Mineral Co. Complete analysis of Zr-4 and oxygen, nitrogen, hydrogen, and carbon determinations in I-Zr were made after testing.

Figure 1. Basal plane pole figures for fine and coarse-grained Zircaloy-4 and for coarse-grained pure zirconium. Each point in (b) and (c) corresponds to the (0002) pole of a single grain.

Mechanical Testing

The plane-strain deformation technique was essentially that described by Ford (16). Specimens were made 8 times as wide as they were thick ($3/4 \times 3/32$ -in. in cross section) to insure plane strain over a large fraction of the gage section (17). They were oriented for extension along the rolling direction and loaded through tungsten carbide indentors aligned in a subpress which was designed to operate at temperatures between 20° and 800°C. The deformation-zone geometry, determined by the indenter-width to sheet-thickness ratio (b/t_0), was varied between 0.9 and 1.7 by changing the indenter width while keeping the sheet thickness constant. The subpress was mounted in a vacuum furnace (18) which in turn was fastened to the crosshead of an Instron testing machine. Reductions were generally made at 100°C intervals at a strain rate of 10^{-1} min^{-1} . Temperature was controlled to $\pm 3^\circ\text{C}$ and continuously monitored with a thermocouple in contact with the specimen. Testing below 300°C was done in air when convection improved the system's thermal response and aided in temperature control. At higher temperatures testing in vacuum prevented oxidation of both the sample and lubricant.

True stress-strain curves were calculated with load-displacement data from the Instron chart record. Friction between the indentors and specimen was minimized by lubricating with molybdenum disulfide. It was found to be an effective lubricant at all temperatures. A

modification of Alexander's technique (19) gave $\mu \approx 0.1$ at 760°C , in agreement with values reported at lower temperatures (20). Stresses were corrected by a friction-hill calculation (21), although this never amounted to more than 7% at yielding or 18% at the largest strain ($\epsilon \approx 0.6$). The true or logarithmic strain, $\epsilon = \ln(t_0/t)$, was based on displacement data from which the elastic deflection of the loading system had been subtracted; a check was made after each test by measuring the impression thickness with a micrometer. An offset of 1% was chosen to define yielding. The short gage length in compression (3/32-in.) made it difficult to measure reliably a lesser strain.

The compression results were supplemented with tensile testing over the same temperature range. Fine-grained Zr-4 sheet specimens with gage sections $1\text{-}1/2 \times 3/8 \times 3/32\text{-in.}$ were pulled in the rolling and transverse directions at a strain rate of $7 \times 10^{-3} \text{ min}^{-1}$. At the higher temperatures a helium atmosphere was used to minimize oxidation. True stress-strain curves were again calculated from load-displacement data. There were small but systematic differences in the behavior of specimens of the two orientations (Fig. 2): Pulling in the rolling direction gave lower values of yield stress, reduction-in-area, and width-to-thickness strain ratio, but the rate of work hardening was generally larger. Plots of $\log \sigma$ vs. $\log \epsilon$ were not lines of constant slope; here, $n = \partial \log \sigma / \partial \log \epsilon$ at selected ϵ . Along the transverse direction, n increased with strain, along the rolling direction it

Figure 2. The uniaxial tensile behavior of fine-grained Zr-4 pulled along the rolling direction (0°, solid points) and transverse direction (90°, open points): (a) Yield stress (0.2% offset) and reduction-in-area at fracture; (b) Strain-hardening exponent ($n = \partial \log \sigma / \partial \log \epsilon$) at $\epsilon = 0.10$; (c) Transverse strain ratios, R and P (measured on broken specimens half way between the fracture surface and grip section).

decreased. Only data from rolling-direction tests are used in comparisons to be made later on.

Microscopy

Surfaces suitable for metallography were prepared by grinding through 600 grit paper, then chemically polishing and etching with a solution of 60 parts HNO_3 , 30 parts H_2O , and 10 parts HF. This treatment removed all scratches and produced an optically active surface under polarized light.

The deformed microstructures and fracture features were examined at the center of the strip in the plane of straining, i.e. that defined by the sheet normal and rolling direction. The twinned volume (fraction of matrix converted to twin orientation) was found by systematic point counting (22) using polarized light to differentiate twin from matrix. A two-surface analysis on grains oriented by the back-reflection Laue technique was used to identify the true orientations of deformation features.

Still higher magnifications were obtained by electron microscopy of carbon replicas. The replicas were made from metallographically polished internal sections by a two-step method, then shadowed at a low angle with germanium. Dislocation structures were studied in Zr-4 by transmission electron microscopy. Thin foils were prepared from deformed specimens by Blackburn's method (23). Scanning electron microscopy was used on the as-fractured surfaces.

RESULTS

Yielding and Flow

The stress-strain curves for Zr-4 in plane-strain compression and uniaxial tension were very much different. In compression the yield stress (σ_y) was about twice that for tension at all temperatures (Fig. 3a). This is higher by about 30% than anything to be calculated from anisotropic plasticity theory (17,24) using the transverse strain ratios, R and P, from tension tests. The compression yield-stress was also affected somewhat by deformation-zone geometry. Lowering b/t_0 from 1.6 to 0.9 raised σ_y by about 10%. Still other differences in work hardening are summarized in Fig. 3b. In compression, the data fit a constant-n curve fairly well. The shorter gage length made these strain measurements less accurate than those for tension, but true stress-strain curves (when they did not end in fracture) could be followed to much larger strains. Above 500°C, however, $\log \sigma$ vs. $\log \epsilon$ plots were no longer straight; now stress reached a constant value or even decreased slightly after a strain of about 0.10. Work hardening in tension as measured by n was greater than in compression.

The stress-strain curves for fine and coarse-grained Zr-4 in plane-strain compression were much alike. The main difference was a 10% higher flow stress in the latter, which was probably caused by

Figure 3. The temperature dependence of the yield stress and strain-hardening exponent in fine-grained Zr-4 under plane-strain compression and uniaxial tension: (a) Yield stress (1% offset) with $b/t_0 = 1.6$; (b) Strain-hardening exponent (in tension for $\epsilon = 0.10$).

its more pronounced (0002) texture. This was in marked contrast to the behavior of pure zirconium (Fig. 4). Below 300°C in I-Zr there was a large scatter in yield stress, load drops on yielding, and higher work-hardening rates. At higher temperatures I-Zr behaved much the same as Zr-4 but it was weaker.

Ductility

The reduction-in-area to fracture is a convenient measure of ductility in tension. In compression, however, the corresponding measurement depends on what constitutes "fracture". Under compressive loading, ductility in Zr-4 was limited by shear cracking which generally began near the indenter corners at the edges of the strip. With continued straining cracking advanced towards the center of the strip until finally the load began to fall. The strain for each such "degree" of cracking was measured, and typical results are shown in Fig. 5. Edge cracking was found by examining at 50X the edges of deformed specimens polished before testing. The same specimens were then sectioned at mid-width, polished, and examined at 100X for internal cracks. The third measure of fracture strain, load falling with increasing strain, came from the Instron chart record. The compressive fracture strains reported later on are those for decreasing load. There is no question but that ductility is lost by then, and these were the easiest measurements to make.

Figure 4. The temperature dependence of yield stress and strain-hardening exponent in coarse-grained Zr-4 and I-Zr under plane-strain compression: (a) Yield stress (1% offset or, in the case of load drops, the stress prior to the drop); (b) Strain-hardening exponent.

Figure 5. Temperature dependence of the strain ($\epsilon = \ln t_0/t$) for various degrees of cracking in fine-grained Zr-4 compressed with $b/t_0 = 1.4$. The points $\circ$, $\square$, and $\diamond$ represent metallographic observations showing no cracks, cracks near the edge only, and cracks across the entire strip, respectively.

It is not too surprising that cracking begins near the indenter corners. For a "square" deformation zone ($b/t_0 = 1$), the slip-line field is a single cross connecting diagonally opposite corners of the zone. Shear tends to concentrate on the planes so defined and fracture develops there first. As the zone takes on non-integral values of b/t_0 , the slip-line field acquires a fan with its apex at the zone corners (25). Strain is now diffused within the fan but is still concentrated at the apex, as on the lines in the first case, and fracture again starts there. The effect of b/t_0 on ductility can be understood at least approximately by letting the boundaries of all fields be straight lines. Shear strain along such a line is then equal to the velocity change parallel to the line, that comes from crossing it, divided by the velocity normal to the line (26). Increasing b/t_0 requires the normal velocity to increase simply because the material is incompressible, but it leaves the parallel velocity change unaffected. Therefore, the shear strain concentration near the indenter corners will be reduced by increasing b/t_0 and the measured ductility should be somewhat greater.

The temperature dependence of the fracture strain, ϵ_f , in both plane-strain compression and uniaxial tension is shown in Fig. 6. In plane-strain compression there was a marked ductility transition at about 300°C. Increasing b/t_0 and temperature gave limited improvement in ductility below 300°C, while above fracturing was no longer a problem. In uniaxial tension, fracture strains were fairly large at 20°C ($\epsilon_f \approx 0.5$) and increased smoothly with temperature to $\epsilon_f \approx \infty$ (rupture) above 600°C.

Figure 6. The temperature dependence of the fracture strain of Zr-4 under plane-strain compression ($\epsilon_f = \ln t_0/t_f$) and uniaxial tension ($\epsilon_f = \ln A_0/A_f$). The insert is a detail of the cross-hatched area illustrating the effect of deformation-zone geometry (b/t_0) on compressive fracture strain. All material is fine-grained except as noted.

The temperature dependence of fracture strain in coarse-grained Zr-4 with $b/t_0 = 1.6$ followed that of the fine-grained material, but with an ϵ_f only half as much. I-Zr compressed with essentially the same b/t_0 never fractured.

Deformation Mechanisms

There were changes in the deformation mechanisms in both I-Zr and Zr-4 just below 300°C. The deformed structures are shown in Fig. 7. Below 300°C, compression produced wide lenticular twins throughout the whole deforming volume in I-Zr. These were followed by second and third-order retwinning within the first twinned volume. The habit plane of the primary twins was $\{11\bar{2}2\}$, in agreement with the findings of Rapperport and Hartley (7). The load drops and large scatter in upper yield point (Fig. 4) were associated with the formation of these twins. Under similar conditions there was very little twinning in Zr-4. The twins which did form were mostly $\{10\bar{1}2\}$, but a few $\{11\bar{2}1\}$ were seen as well; both give c-axis extension. They occurred primarily in grains out of the (0002) texture, oriented with c-axes close to the transverse direction. It was not uncommon for grains of that orientation to be 50 to 90% twinned. Although two conjugate planes were usually active in a given grain --- $(10\bar{1}2)$ and $(\bar{1}012)$ --- the 86° reorientation associated with each rotated the twinned volumes in the opposite sense to nearly the same orientation, which is in the (0002) texture. During further reduction these grains behaved

Figure 7. Typical microstructures produced in I-Zr and Zr-4 deformed in plane-strain compression above and below 300°C: (a) I-Zr compressed to $\epsilon = 0.15$ at 200°C; (b) Zr-4 compressed to $\epsilon = 0.15$ at 200°C; (c) I-Zr compressed to $\epsilon = 0.18$ at 400°C; (d) Zr-4 compressed to $\epsilon = 0.14$ at 400°C. The diagonal in the upper right-hand corner is the trace of the sheet surface.

the same as those initially of texture orientation. Occasionally small $\{10\bar{1}2\}$ twins were seen in grains compressed along $[0001]$. These must have formed on unloading to relieve residual stresses. At and above 300°C , I-Zr and Zr-4 were very similar. There was considerable twinning but it was now on $\{10\bar{1}1\}$. The appearance of these twins was distinctive. They were always quite narrow and tended to form in tight packets made up of many parallel twins. With increasing strain the fraction twinned within a packet increased until occasionally second-order twins formed within consolidated packets.

The behavior of the I-Zr was nearly identical to that reported by Paton for titanium of similar purity (27). The only real difference was a 100°C higher temperature for the twin-transition from $\{11\bar{2}2\}$ to $\{10\bar{1}1\}$ in the titanium.

The strain accommodated by twinning can be calculated from twinned volume measurements if the twin plane and shear are known (28) and an ideal (0002) texture is assumed. Figure 8 shows the strain dependence of twinning at 20°C . The broken line represents the volume fraction required if all strain were from $\{11\bar{2}2\}$ twinning. A fully twinned matrix would give a compressive strain of 0.110 (Table 2). I-Zr follows this line through $\epsilon \approx 0.04$, then bends away with increasing strain. The bend corresponds with the beginning of second and third-order retwinning. Hobson has identified these higher-order twins as $\{10\bar{1}2\}$ and $\{11\bar{2}1\}$ from the development of texture in a single crystal compressed along $[0001]$ at

Figure 8. Volume fraction twinned (V_t) vs. strain ($\epsilon = \ln t_0/t$) for I-Zr and Zr-4 deformed in plane-strain compression at 20°C. 95% confidence limits are shown.

TABLE 2
TWINNING SHEARS

K_1 Habit Plane	S Twin Shear Strain	ϵ_c Principle Strain ($\ln t/t_0$) Along Original G-axis*
$\{10\bar{1}1\}$	0.104 [†]	- 0.045
$\{10\bar{1}2\}$	0.167 ⁽⁷⁾	+ 0.083
$\{11\bar{2}1\}$	0.627 ⁽²⁹⁾	+ 0.167
$\{11\bar{2}2\}$	0.226 ⁽⁷⁾	- 0.110
$\{11\bar{2}3\}$	unknown	----

* (+) for extension and (-) for compression.

† The $\{10\bar{1}1\}$ twinning shear has not been measured in Zr. It was calculated assuming the same twin elements as those reported in Ti (27).

20°C (30). No quantitative measure of higher-order twinning was possible here, but it was apparent from the microstructure (Fig. 7a) that most of the primary twins had retwinned at least once. The strain in I-Zr can be accounted for by twinning alone if shears associated with higher-order twins are included. In Zr-4 the few $\{10\bar{1}2\}$ and $\{11\bar{2}1\}$ twins did not contribute to c-axis compression.

The temperature dependence of twinning is summarized in Fig. 9. Through 200°C, twinning remained the primary deformation mode in I-Zr, while over the same temperature range it produced no c-axis reduction in Zr-4. At higher temperatures, both materials twinned extensively on $\{10\bar{1}1\}$, but the shear on this system is low. In a fully twinned crystal the compressive strain along the c-axis is only 0.045. At all temperatures, $\{10\bar{1}1\}$ twinning never accounted for more than 20% of the imposed strain. Above 500°C, annealing of the microstructures negated twinned-volume measurements even though the specimens were cooled within 30 seconds after straining. Twinning did persist, however, through at least 700°C. At 800°C the specimens could not be cooled quickly enough to prevent complete recrystallization.

Strain not accounted for by twinning must, of course, be based on slip. Slip, then, was the primary deformation mechanism in Zr-4 at all temperatures and in I-Zr at 300°C and above. Reductions along the c-axis can only be accomplished by slip on systems with a $\langle c \rangle$ component in their Burgers vector. The kink bands shown in Fig. 10

Figure 9. Volume fraction twinned (V_t) vs. temperature for a selected $\epsilon = 0.15$. At each temperature a specimen was generally strained to $\epsilon = 0.15 \pm 0.03$. The values shown here along with their 95% confidence limits were found by extrapolating volume fraction twinned vs. strain curves (such as that in Fig. 8) to $\epsilon = 0.15$.

Figure 10. Polarized light micrograph of kink bands formed in Zr-4 during compression at 20°C. The surface is an internal section prepared after straining. Contrast is provided solely by changes in basal-plane orientation.

are evidence for such slip. The bands were inclined anywhere from 10° to 40° (true angle) to the basal plane. If made up of an array of edge dislocations (31), they must have formed by slip on $\langle c + a \rangle$ systems. Kinks of the type shown in Fig. 10 were seen in specimens compressed at temperatures from 20° to 100°C . At higher temperatures similar changes in basal-plane orientation occurred, but the boundaries were more gradually bent than sharply kinked.

Since the textures in these materials were not perfect (0002), some slip on systems with $\langle a \rangle$ type Burgers vectors was expected. Differences in the character of $\langle a \rangle$ and $\langle c + a \rangle$ type slip appear to be of prime importance to ductility. The transmission micrographs of Fig. 11 illustrate these differences. Dislocations with $\langle a \rangle$ Burgers vectors (from prism, basal, or pyramidal slip) were seen to some extent in most grains. They were always more or less uniformly distributed throughout the matrix as in Fig. 11a. On the other hand, $\langle c + a \rangle$ slip occurred in fewer grains with the dislocations confined to thin bands of the type shown in Fig. 11c. Typically these bands were 0.5 to 2 μm thick and spaced about 10 μm apart. The orientation of the bands, from a trace analysis (32) based on electron diffraction patterns, was close to $\{11\bar{2}4\}$.

The Fracturing Process

Bands of highly localized shear developed in Zr-4 compressed at temperatures below the ductility transition. Typical examples in both the fine and coarse-grained materials are shown in Fig. 12. Slight

Figure 11. Transmission micrographs of Zr-4 compressed to $\epsilon = 0.012$ at 20°C . The zone axes are $\langle 11\bar{2}0 \rangle$: (a) $g = 10\bar{1}1$; (b) Same area as (a) with $g = 0002$. The dislocations in (a) have $\langle a \rangle$ type Burgers vectors. (c) $g = 0002$; dislocations in contrast have $\langle c + a \rangle$ type Burgers vectors.

Figure 12. Shear bands formed in Zr-4 during plane-strain compression: (a) Electron micrograph from replica of fine-grained material compressed to $\epsilon = 0.33$ at 100°C ; (b) polarized light micrograph of coarse-grained material compressed to $\epsilon = 0.61$ at 200°C .

preferential attack by the chemical polish made the bands readily visible, even on internal sections. The large shear displacements along them were evident from steps produced in grain boundaries where bands crossed from one grain into the next. The greatest difficulty in finding the strain within a shear band was in measuring its thickness, which was never more than about 1 μm . Grain boundary offsets of 50 to 100 μm were not uncommon, giving shear strains in excess of 50. Continued shear within these bands eventually led to cracking.

The orientations of a number of shear bands along with the orientations of the grains in which they formed are assembled in Fig. 13. The bands developed in grains compressed along [0001], or in grains which could only deform by $\langle c + a \rangle$ slip. They were not, however, simply a product of the strain concentration required by the plane-strain technique. Were that the case, they should have formed only near the indenter corners and been inclined only toward the rolling direction. Instead they were found throughout the whole deforming volume (particularly in the coarse-grained material) and were often inclined away from the plane of strain (Fig. 13b). This observation was further confirmed by compressing a few rectangular specimens of Zr-4 without the constraint of plane-strain, and they also developed shear bands. The bands appeared, therefore, to have a basic structural, i.e. crystallographic, origin. The band normals, when plotted on a standard stereographic triangle, were closely grouped near the $\{11\bar{2}4\}$ pole (Fig. 13c). The formation of shear bands and $\langle c + a \rangle$ dislocation

Figure 13. Shear banding in plane-strain compression: (a) Orientation of the basal plane in grains which developed shear bands; (b) Orientation of shear bands relative to sheet axes; (c) Orientation of shear bands relative to crystal axes.

bands in grains of the same orientation and along planes with the same indices supports the idea that the former develop from the latter. The structural problem then becomes one of explaining continued $\langle c + a \rangle$ dislocation glide along $\{11\bar{2}4\}$ planes. The strain concentration in plane strain merely encourages this development.

Figure 14 illustrates what can happen when a $\{11\bar{2}4\}$ plane is aligned with the indenter edge and inclined at about 45° (here 40°) to the plane of the sheet. A shear band started at that edge, then propagated through the first and second grains. The second grain offered little resistance to the band, whereas the third was a much stronger barrier. The effectiveness of a given grain in arresting a shear band depended on the change in orientation across the boundary being penetrated. During the texture measurements it was not uncommon to find four consecutive grains oriented within 10° of each other. This accounts for the high frequency with which these crystallographic bands were able to cross from one grain into the next.

The actual separation process along a band was that of most ductile fractures: the nucleation, growth, and coalescence of voids. Fracturing at 20°C was particularly violent, sending the overhanging portions of the specimen flying. Surfaces on pieces separated in this way were suitable for fractography. Approximately 30% of the fracture area was covered with parabolic dimples (Fig. 15) elongated in the proper sense on mating surfaces for hole growth in shear (33). The remainder was

Figure 14. Shear band formed in coarse-grained Zr-4 compressed to $\epsilon = 0.15$ at 200°C . The insert at the upper left shows the area of the micrograph.

Figure 15. Scanning electron micrographs of fracture surfaces produced by plane-strain compression of Zr-4 at 20°C.

rather smooth and featureless, probably because of rubbing before complete separation. At higher temperatures, 100° to 300°C, surfaces were totally obscured by rubbing.

DISCUSSION

In compression limited ductility was found only in Zr-4 at temperatures below 300°C. The texture and loading system demanded reduction along the c-axis; the material and temperature allowed only $\langle c + a \rangle$ slip. Under these conditions shear became localized to the point of fracture.

In I-Zr under the same conditions, $\{11\bar{2}2\}$ twinning acted to distribute the strain more or less uniformly throughout the whole deforming volume (Fig. 7a). There appeared to be a minimum applied stress for forming these twins and this stress decreased with falling temperature (along the lower curve in Fig. 4) suggesting that $\{11\bar{2}2\}$ twin nucleation is essentially a mechanical process. Twinning on $\{11\bar{2}2\}$ in I-Zr stopped somewhere between 200° and 300°C, the temperature range in which the yield stress for slip dropped below that for twinning. The absence of this twin mode in Zr-4 implies that its critical stress exceeds that for slip over the whole test-range of temperature.

The formation of $\{10\bar{1}1\}$ twins followed quite another pattern. Twinning on these planes occurred in both materials at and above 300°C. The lower limit on the twinning temperature, even though the yield

stress in Zr-4 increases rapidly with decreasing temperature, would seem to mean that there is a barrier in the nucleation of these twins which must be overcome by thermal activation.

The large minimum-stress for $\{11\bar{2}2\}$ twinning and the apparently thermally activated nature of $\{10\bar{1}1\}$ twinning leave Zr-4 without a compressive twin mode below 300°C. Slip on systems with $\langle c + a \rangle$ Burgers vectors has been established with transmission microscopy, but the problem remains of why shear on these systems should become so localized. Such slip is confined to bands (Fig. 11c) and a number of these eventually develop very large shear displacements (Fig. 12). Continued shear within the bands suggests that any strain hardening there is being overshadowed by other factors acting to localize flow. One possibility, of course, is softening from heat generated during slip-band formation.

To explore that possibility requires first estimating the fraction of heat, α , which must be retained within a deforming volume if thermal softening is to overcome strain hardening. This quantity can then be compared with the fraction of heat, α' , retained during slip-band formation which, in principle, is calculable from standard heat-flow analysis if details about band size and formation time are known. If $\alpha \leq \alpha'$, flow ought to continue within the band, even to the point of fracture.

If strain-rate effects are discounted, the condition for thermal softening is

$$\frac{d\sigma}{d\epsilon} = \left. \frac{\partial\sigma}{\partial\epsilon} \right|_T + \left. \frac{\partial\sigma}{\partial T} \right|_{\epsilon} \frac{dT}{d\epsilon} \leq 0 \quad (1)$$

Strain hardening in compression was reasonably well described by $\sigma = K\epsilon^n$ with n being essentially independent of temperature (Figs. 3 and 4); therefore $(\partial\sigma/\partial\epsilon) \equiv n\sigma/\epsilon$ and $(\partial\sigma/\partial T) \equiv (\partial K/\partial T)\epsilon^n$. With α the fraction of deformation work retained as heat, $\alpha\sigma d\epsilon = \rho c dt$, in which ρ is density and c is heat capacity. Then Eq. (1) becomes

$$\frac{d\sigma}{d\epsilon} = \sigma \left[\frac{n}{\epsilon} + \frac{\alpha\epsilon^n}{\rho c} \frac{\partial K}{\partial T} \right] \leq 0 \quad (2)$$

Equation (2) is satisfied when the quantity in brackets is ≤ 0 , which is the basis for finding α .

Now consider α' , the fraction of heat retained within a slip band in which the heat generation rate is constant during a lifetime, t . The band is assumed to have a constant thickness, w , through all of t . With heat loss by conduction to the surrounding matrix, α' at the center of the band is (34)

$$\alpha' = \left[1 - 4i^2 \operatorname{erfc} \frac{w}{4\sqrt{\kappa t}} \right] \quad (3)$$

in which $i^2 \operatorname{erfc}$ is the double integral of the complimentary error function and κ is thermal diffusivity.

The next step is to equate α from the bracketed quantity in Eq. (2) to α' from Eq. (3). Doing this presupposes that strain hardening in the slip band can be represented by what is measured with a test piece. The result is

$$\frac{\frac{n}{\partial T} \frac{\rho c}{\epsilon (n+1)}}{\partial T} = \left[1 - 4i^2 \operatorname{erfc} \frac{w}{4\sqrt{\kappa t}} \right] \quad (4)$$

Values for n and K were taken from $\log \sigma$ vs. $\log \epsilon$ curves at various temperatures, and these showed $n = 0.08$ (Figs. 3 and 4) and $\frac{\partial K}{\partial T} \approx -0.24 \text{ kg/mm}^2/\text{°C}$. Other data are $\rho = 6.49 \text{ gm/cm}^3$, $c = 0.066 \text{ cal/gm/°C}$, and $\kappa = 0.070 \text{ cm}^2/\text{sec}$ (35). Three factors remain to be evaluated: the band width, w , the strain within a band, ϵ , and the time of band formation, t . The most elusive, yet most critical, of these is t , and it has only recently been measured by Fisher and Lally (36,37). Their conclusion, reached from studies of acoustic emission and micro load-drops was that deformation occurs discontinuously by slip avalanches which last no longer than a few microseconds, regardless of the imposed strain rate. At least seven different materials were tested and in each case t was found to be about $5 \mu \text{ sec}$. Using this value, Eq. (4) gives the combinations of band width and band strain at which flow should become unstable, these are plotted for Zr-4

in Fig. 16. A combination of properties acts to locate the curve in Fig. 16 as close to the coordinates as it is: a flow stress which falls rapidly with increasing temperature (Figs. 3 and 4), a low strain-hardening rate, and, to a lesser extent, low thermal diffusivity. Such an analysis for BCC metals gives similar results only below about 50°K, where flow stress becomes strongly temperature dependent and strain-hardening rate decreases rapidly (38). For FCC metals, this idea is most useful near absolute zero where it is the low heat capacity which favors thermal softening (39).

Slip bands as thick as 2 μm were seen with transmission microscopy. From Fig. 16, the corresponding instability strain is $\epsilon \approx 0.4$ (or a shear strain of $\gamma \approx 0.8$), which seems reasonable to expect in at least a few bands of this size. Shear within these bands should then continue. In some, the shear might be arrested at obstacles such as grain boundaries. Whatever the details, however, "damage" can be expected within the bands as voids nucleate and grow around precipitate particles. Fracture follows when enough of these weakened shear bands are linked together.

Above 300°C fracturing from localized shear was not a problem even though $\langle c + a \rangle$ slip is still a primary deformation mechanism in Zr-4 and becomes one in I-Zr. The temperature dependence of the flow stress (Figs. 3 and 4) begins to decrease at that temperature, thereby decreasing the tendency for thermal softening. A few bands of localized shear were seen, however, in specimens compressed between 300° and 400°C. In these cases, twinning on $\{10\bar{1}1\}$, which starts just below 300°C (Fig. 9),

Figure 16. Values of band thickness, w , and band strain, ε , which satisfy Eq. (4).

appeared to blunt the bands (Fig. 17). Shear within the bands was restricted and the imposed strain was more uniformly distributed,

Fracture strains in tension were always large (Fig. 6) because shear did not become so highly localized. Deformation under tensile loading occurs primarily by prism slip (4-6); $\langle c + a \rangle$ slip is not required except, perhaps, for maintaining compatibility at grain boundaries. In fact it is difficult to suppress prism slip when stresses are applied only in the plane of the sheet. This was demonstrated by the behavior of a plane-strain tension specimen designed to restrict flow in the same sense as in the compression tests. Rather than thinning through the sheet thickness, the specimen developed channels along intersecting prism planes and the material between necked to a knife edge. A fracture surface produced in this way is shown in Fig. 18.

Figure 17. Shear bands formed in Zr-4 under plane-strain compression: (a) Compressed to $\epsilon = 0.625$ at 300°C; (b) Compressed to $\epsilon = 0.625$ at 350°C.

Figure 18. Fracture surface of a plane-strain tension specimen pulled at 20°C: (a) overall view; (b) side view of facets produced by prism slip; (c) overhead view of knife edge produced by ductile rupture.

SUMMARY AND CONCLUSIONS

Zircaloy-4 sheet with strong (0002) texture undergoes a transition in ductility at about 300°C when it is compressed through the sheet thickness in plane strain. Below this temperature strains are limited by shear cracking; above, large fracture-free reductions can be made. Pure zirconium under identical conditions is ductile at all temperatures.

The differences in ductility are a reflection of the way compressive strains along the c-axis are accommodated. Zircaloy-4, lacking a compressive twin mode below 300°C, slips on systems with $\langle c + a \rangle$ Burgers vectors. Dislocations of this type are confined to thin bands oriented close to $\{11\bar{2}4\}$. Over the same temperature range deformation in pure zirconium is by $\{11\bar{2}2\}$ twinning followed by second and third-order re-twinning within the primary twins. At 300°C and above both deform by a combination of $\{10\bar{1}1\}$ twinning and $\langle c + a \rangle$ slip.

Fracturing in Zircaloy-4 is by highly localized shear along bands of $\{11\bar{2}4\}$ orientation. Continued shear within bands heavily populated with $\langle c + a \rangle$ dislocations suggests that strain hardening there is being overcome by other factors acting to localize flow. An analysis is developed which suggests that heat generated during slip-band formation can soften a band sufficiently to make shear continue there.

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