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RADIATION DAMAGE IN HEXAGONAL CLOSE-PACKED METALS AND ALLOYS

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ABSTRACT

The effects of radiation damage to hcp metals and alloys are discussed, based upon a comprehensive investigation of Zircaloy-2 conducted at Battelle-Northwest. Irradiations were conducted at both 60 and 280 C to exposures extending to 2×10^{21} n/cm². Tensile properties were measured for transverse and longitudinal specimens taken from cold-rolled plate containing various levels of cold work. The effects of radiation damage to plastic flow and fracture behavior varied greatly with both irradiation and testing temperature. At exposures greater than about 5×10^{20} n/cm², damage at 280 C was greater than at 60 C. Even though changes in yield strength with neutron dose at 280 C fit a saturation model, saturation did not occur at the highest exposure obtained. The effects of crystallographic anisotropy are preserved and in some respects intensified by neutron damage. At high neutron doses at 280 C, yield strength becomes nearly independent of cold work indicating that radiation-induced hardening overrides strain-induced hardening. The above effects are discussed with reference to basic deformation and fracture mechanisms applicable to a variety of hcp metals and alloys.

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1.0 INTRODUCTION

The hcp metals and alloys form an interesting family of materials for irradiation damage studies because of their specific deformation mechanisms and inherent anisotropy. Some members of this family such as zirconium, beryllium, and magnesium and their alloys have application in nuclear reactors and for fuel cladding because of their unique nuclear properties. In spite of this, these metals have received relatively little attention in basic irradiation damage studies in comparison with bcc and fcc metals. Only a few studies have set out to show the effect of crystallographic structure on the accumulation and recovery of radiation damage.

The bulk of the irradiation investigations of hexagonal metals and alloys have been conducted on zirconium and the Zircaloy alloys because of their widespread use in hot-water reactor systems. The principal goal of most of these studies has been to assess the effects of irradiation on the mechanical behavior of structural components. There is also considerable interest in radiation effects to corrosion behavior and pickup of corrosion hydrogen. Effects of hydrogen embrittlement are reviewed elsewhere,^(1,2,3) and are discussed only briefly in this paper. Most of the data presented are taken from a comprehensive study of irradiation effects to Zircaloy-2 being conducted at Battelle-Northwest under sponsorship of the Division of Reactor Development and Technology, USAEC.

2.0 UNIRRADIATED PROPERTIES

2.1 Crystallographic Textures

The plastic deformation of polycrystalline Zircaloy-2 alters the lattice orientation of individual grains so that certain lattice directions develop a preferred orientation with respect to the principal directions of flow. These preferred orientations are not eliminated by recrystallization, but are rotated into new orientations.⁽⁴⁾ This is illustrated in Figure 1 where pole figures for the principal lattice planes in annealed and 70% cold-worked Zircaloy-2 are presented.⁽⁵⁾ The cold-rolled texture is one in which the (0002) basal planes contain the rolling direction and are distributed about the rolling direction in a fiber-like texture. The predominant orientation of the hexagonal unit cell is normal to the rolling plane with the (10 $\bar{1}$ 0) prism planes normal to the rolling direction.

After a recrystallization treatment the basal plane orientation is not greatly altered, but the hexagonal unit cell rotates nearly 30° about the C-axis, which exchanges the positions of the (10 $\bar{1}$ 0) and (11 $\bar{2}$ 0) prism planes.^(6,7) After this rotation a definite orientation of (10 $\bar{1}$ 0) poles is retained in the rolling direction.

The textures developed in Zircaloy-2 by cold work and annealing are similar to those for zirconium. Similarities also exist in the textures for sheet and tubing. In all cases, the basal planes are oriented approximately 90 degrees from the working direction. Differences are in the distribution of the basal poles between the other two principal directions and the degree of alignment of $\langle 10\bar{1}0 \rangle$ and $\langle 11\bar{2}0 \rangle$ directions in the working direction.

2.2 Plastic Flow Behavior

For hcp metals with a c/a ratio less than 1.633 (for perfect spheres), it can be shown that the interplanar spacing and atomic density of the prismatic $\{10\bar{1}0\}$ planes are greater than those for the basal planes. This is because the lattice is compressed along the c -axis, reducing the interplanar spacing and atomic density of the basal planes. Rapperport⁽⁸⁾ and Rapperport and Hartley⁽⁹⁾ in their studies of deformation in zirconium observed only $\{10\bar{1}0\} \langle \bar{1}2\bar{1}0 \rangle$ slip. The active twin planes were $\{10\bar{1}2\}$ and planes of the form $\{11\bar{2}X\}$, ($X = 1, 2, 3$). Twins of the form $\{10\bar{1}2\}$ and $\{11\bar{2}1\}$ were observed when tension was applied in the direction of the basal pole, and twins of the form $\{11\bar{2}1\}$ and $\{11\bar{2}2\}$ were observed when compression was applied. The frequency of twins at 300 K in descending order was $\{11\bar{2}1\}$, $\{10\bar{1}2\}$, $\{11\bar{2}2\}$ and $\{11\bar{2}3\}$, the last form being rare.

The $\{10\bar{1}2\}$ and $\{11\bar{2}2\}$ modes of twinning can be described in terms of relatively simple dislocation models.⁽¹⁰⁾ For the $\{11\bar{2}1\}$ mode of twinning, however, the model is very complex. These models are in accord with the dislocation theory of twinning given by Cottrell and Bilby⁽¹¹⁾ and by Thompson and Millard,⁽¹²⁾ and they satisfy Cahn's rule relating twin thickness and shear strain.⁽¹³⁾ Homogeneous shear occurs on only every second, every third, and every seventeenth composition plane for $\{10\bar{1}2\}$, $\{11\bar{2}2\}$, and $\{11\bar{2}1\}$ twinning, respectively. The models show that twinning is nucleated at stress concentrations, so that dislocation pile-ups due to prior slip probably precedes twin nucleation in polycrystalline material.⁽¹⁰⁾

The modes of deformation twinning in Zircaloy-2 are the same as those for zirconium,⁽⁶⁾ except that only $\{10\bar{1}2\}$ twinning appears to be active during tensile strain at room temperature. Both $\{10\bar{1}2\}$ and $\{11\bar{2}1\}$ twinning are important deformation modes at liquid nitrogen temperature (77 K), however.⁽¹⁴⁾ Deformation at this temperature favors twin nucleation but not growth. Twins are found in a much larger fraction of the grains, and the average number of twins per grain is about an order of magnitude greater than at room temperature. Cross-twinning and second-order twinning, which are generally not observed in zirconium deformed at room temperature, are observed with tensile deformation at 77 K.⁽¹⁴⁾ The incidence of twinning in both zirconium and Zircaloy-2 strained in tension at 540 C (813 K) is much reduced.

Martin and Reed-Hill⁽¹⁵⁾ found that kink bands involving basal slip are an important mode of deformation in polycrystalline zirconium. The relative amount of kinking increases with deformation temperature due to the contribution of dislocation climb in the alignment of kink-band boundaries. Kink bands occur primarily in grains having the basal planes inclined greater than about 30° from the tensile axis. Since grains having this orientation usually deform by twinning, slip kinking and mechanical twinning may be considered competitive deformation processes.

The strong preferred orientations developed in Zircaloy-2 and the influence of these crystallographic textures on deformation mechanisms cause anisotropy in mechanical properties. The nature of this anisotropy is illustrated in Figure 2.⁽¹⁶⁾ In the annealed condition,

yielding is continuous in the longitudinal direction and discontinuous in the transverse direction. This latter form of yielding accompanies the establishment of additional deformation elements by twinning and does not involve a drop in load.

Transverse specimens strain harden beyond the short, initial yield region, but at a lower rate than for longitudinal specimens. This reduced rate of strain hardening has a pronounced effect on tensile behavior. The stress-strain curves for annealed Zircaloy-2 cross at about 3% strain, and the flow stress at larger strains is higher in the longitudinal direction. Consistent with this crossover for annealed material, the stress-strain curves for longitudinal specimens lie above those for transverse specimens at each of the various cold work levels. Other effects stemming from this directional variation in strain hardenability include a lower uniform elongation, lower tensile strength, higher necking strain, and higher reduction of area in transverse compared to longitudinal specimens.

Figure 2 shows that uniform elongation decreases sharply between 0 and 10% cold work, then increases slightly with higher levels of cold work. This retroflexion is attributed to the development of new deformation systems which contribute to strain hardening as plastic deformation progresses.

Since slip in Zircaloy-2 occurs principally on prism planes, slip is promoted when the tensile axis lies in the basal plane such as occurs in longitudinal specimens. When the stress axis is normal to the basal plane, none of the prism planes are favorably aligned for slip. Thus, slip in transverse specimens occurs readily only for the

few grains having basal planes aligned in the rolling plane. Grain rotation or twinning is required in the remaining grains to bring additional prism planes into a more suitable alignment for slip.

Figure 3 illustrates the appearance of both longitudinal and transverse specimens in the annealed condition after 10% plastic strain in tension. Deformation in the longitudinal specimen is almost entirely by slip, whereas deformation twins of the $\{10\bar{1}2\}$ mode are prominent in the transverse specimen. For a given amount of strain, twinning in transverse specimens is about a factor of ten more abundant than in longitudinal specimens. However, even in the transverse direction, twinning accounts for only about 10% of the total strain. ⁽¹⁴⁾

2.3 Yield Behavior

The yield phenomenon in hcp metals and alloys is not completely understood. Keeler⁽¹⁷⁾ observed a grain size dependency in the occurrence of a sharp yield point in zirconium. Small-grained material (grain diameter of 0.003 mm) exhibited a yield point over the range -195 C to 300 C, whereas specimens having a larger ^{grain size} ~~yield point~~ did not exhibit a yield point at any temperature. Grain size is not the only controlling variable, however, since Westlake⁽¹⁸⁾ has observed yield points in single crystals of zirconium.

In Keeler's investigation,⁽¹⁷⁾ yielding was sharper at higher than lower temperatures; and in studies by Shober and co-workers⁽¹⁹⁾ on Zircaloy-2 containing low interstitial content, yield points occurred only at temperatures above about 230 C. Although attempts have been made to attribute discontinuous yielding in these materials to a Cottrell or Suzuki locking mechanism involving interstitials, all

observations are not explained by these mechanisms nor are all classical tests for these mechanisms confirmed by the observations.

A more recent yield-point model advanced by Johnston⁽²⁰⁾ and Hahn⁽²¹⁾ and based on dislocation mechanics is gaining acceptance.

The occurrence of an abrupt yield point by this model is based upon three requirements; namely, (1) the number of initially mobile dislocations is small, (2) dislocations multiply rapidly at a stress corresponding to the upper yield point, and (3) dislocation velocity is dependent upon the applied stress. The magnitude of the yield drop is increased the greater the multiplication rate of mobile dislocations and the higher the effect of dislocation velocity on stress (or conversely, the lower the effect of stress on dislocation velocity).

Among the possible means for promoting a yield point by pinning potentially mobile dislocations are impedance by dislocation sub-boundaries, precipitation of interstitial impurities, and agglomeration of point defects. Directional variations in yield phenomena should occur due to the variation in possible slip systems and, hence, the density of mobile dislocations with test direction. Prior cold work reduces the potential for a yield point by greatly increasing the density of mobile dislocations.

A yield point by the Hahn model requires a multiplication mechanism such as double cross-glide in addition to a pinning mechanism. Such a mechanism in zirconium or Zircaloy-2 could occur by cross-glide of screw dislocations from prism to basal planes, which share a common slip direction. Since cross-glide is thermally activated, dislocation multiplication would be greater the higher the temperature. Furthermore,

from the observation by Martin and Reed-Hill⁽¹⁵⁾ that basal glide is favored in the transverse direction, it follows that dislocation multiplication should also be more prevalent in the transverse than in the longitudinal direction.

2.4 Recovery Behavior

The effects of cold work recovery during thermal annealing treatments have an important bearing on the stability of irradiated Zircaloy-2. Plots of yield strength and uniform elongation as functions of cold work both before and after annealing treatments at 360 and 425 C are shown in Figure 4.⁽²²⁾ Whereas recovery is substantial in the longitudinal direction at both annealing temperatures, significant recovery in the transverse direction occurs only at 425 C. The lower response of transverse specimens to thermal recovery is attributed to higher substructure restraints^t on deformation in this direction. The applied flow stress for twinning, which requires stress concentrations for nucleation, is less affected by internal stress recovery than is the applied stress for slip.

Directional variations in recovery affect the anisotropy in tensile properties for cold-worked specimens. Longitudinal specimens, which have a higher yield strength than transverse specimens before annealing, have a lower yield strength after annealing. Furthermore, the anisotropy in both uniform and total elongation increases markedly with annealing temperature.

2.5 Fracture Behavior

The mechanisms for ductile fracture observed in Zircaloy-2 in the absence of hydrogen are essentially those proposed by Rogers for

copper.⁽²³⁾ Both longitudinal and transverse specimens display a shear lip connecting the internal portion of the tensile break to the outer surface of the specimen. The amount of shear lip increases with cold work for both longitudinal and transverse specimens; however, the proportion of shear fracture in transverse specimens is greater. Generally, the amount of shear fracture is inversely related to strain hardenability.

Pore formation is a preliminary step to the initiation and propagation of ductile fracture.⁽⁵⁾ Generally, the void-containing region is smaller for transverse than for longitudinal specimens, decreases with increasing cold work, and is more prominently associated with the internal break than with the shear lip. The voids are generally nucleated as micro-fissures at inclusions, hydrides, or other second-phase particles, and in some instances the particle is cleft with the matrix. Stress concentrations at the ends of the fissures are relaxed by plastic deformation allowing them to open up in the tensile direction as diamond-shaped voids.

Plastic deformation during the necking process is strongly influenced by crystallographic anisotropy.⁽⁵⁾ The transverse and normal directions with respect to rolling are oriented, respectively, in the width and thickness directions of longitudinal specimens. The intensity of basal poles is strong about the tensile axis, but is strongest in the thickness direction. This orientation favors necking in the width direction, where contractile strain can occur normal to the basal pole. Because grains having basal poles oriented in the width direction cannot contribute to this strain, the neck spreads over a

relatively wide zone along the tensile axis. Lateral restraints imposed by such a neck develop a state of triaxial tensile stress and favor widespread fissuring and void formation.

The width and thickness directions of transverse specimens coincide with the rolling and normal directions, respectively. A preponderance of grains are oriented with basal poles in the thickness direction, but only a few with basal poles in the width direction. Not only is contractile strain in the width direction highly favored, but also the majority of the grains contribute to this strain. The result is a highly localized neck favoring shear along planes of maximum resolved shear stress. Necking continues until fracture occurs at the center of the specimen and spreads to the zones of macroscopic shear deformation; then, the fracture spreads to the surface along these zones. This type of fracture corresponds to the type proposed by Orowan for nonstrain-hardening materials.⁽²⁴⁾

3.0 IRRADIATION EFFECTS

3.1 Low-Temperature Irradiation

Conventional stress-strain curves for annealed Zircaloy-2 tested in tension at room temperature before and after neutron irradiation at 60 C are presented in Figure 5. Irradiation substantially increases yield strength for both longitudinal and transverse specimens to levels equivalent to 5 and 20% cold work at 0.84×10^{19} n/cm² and 9 and 30% cold work at 1.1×10^{20} n/cm², respectively. In unirradiated material, longitudinal yield strength is greater than transverse yield strength above 3% cold work. In irradiated material, however, transverse yield strength is the greater at each exposure level. Hence, any analogy

between strain hardening and irradiation hardening falls down on two counts: nonequivalence in both the magnitude and relationship of yield strength with stress direction.

In general, the effects of directionality in the plastic behavior of annealed Zircaloy-2 is greater after irradiation than before, and increases with exposure. This is particularly true for both the strain to maximum load and the strain to fracture, as shown in Figure 5. A drop-in-load yield point develops in the transverse direction with exposure to 0.84×10^{19} n/cm² and becomes more acute with exposures to 1.1×10^{20} n/cm² and 2.3×10^{21} n/cm². Continuous strain hardening occurs with yielding in the longitudinal direction after all three exposures.

Twinning occurs in irradiated Zircaloy-2 much as it does in the unirradiated state. Furthermore, the marked variation in twinning with stress direction is preserved. Fracture processes are also relatively unchanged in annealed Zircaloy-2 irradiated at 60 C. However, a somewhat larger shear lip occurs in transverse specimens due to reduced strain hardenability. This increased influence of plastic shear in the fracture is evident from the linear stress-strain relationship beyond yielding at high neutron exposure. Other characteristics of ductile fracture not greatly affected by irradiation to 2.3×10^{21} n/cm² are the reduction of area, the true fracture stress, and the nature of void formation. Although fracture mechanisms are little changed, work expended in the rupture of transverse and longitudinal specimens is reduced by a factor of 7 and 3, respectively, with irradiation to 2.3×10^{21} n/cm².

3.2 Elevated Temperature Irradiation

Conventional stress-strain curves for Zircaloy-2 specimens irradiated at 280 C are presented in Figure 6. These curves are similar to those for 60 C irradiations, except for important differences in the yield behavior and in the magnitude of both flow stress and total strain after large doses. These differences are best described by considering strength levels and anisotropy relationships within three exposure zones.

Stress-strain curves representing the lowest doses obtained at 60 C (0.84×10^{19} n/cm²) and 280 C (5.0×10^{19} n/cm²) are similar despite the much higher dose at 280 C. Except for a lower total elongation at 280 C, flow stresses, yield behavior, and yield strength anisotropy are comparable for both irradiation temperatures.

Irradiation temperature effects at higher exposure are seen by comparing specimens 5-2 and 6-1 in Figure 6 with samples A5 and B5 in Figure 5. Although samples 6-1 and B5 (representing the longitudinal direction) exhibit comparable values of yield strength and tensile strength, samples 5-2 and A5 (representing the transverse direction) do not. Not only is the yield strength in the transverse direction much greater after 280 C irradiation (91,200 psi versus 77,500 psi), but also the yielding appearance is different. Yielding in sample 5-2 occurs over a broad region characteristic of irradiated, cold-worked specimens; whereas sample A5 exhibits a distinct yield point. This marked anisotropy in yield strength after 280 C irradiation to 1×10^{20} n/cm² characterizes a second exposure zone. Again, total elongation in the longitudinal direction is considerably less at the higher

irradiation temperature, but does not differ much with temperature in the transverse direction.

A third type of response to 280 C irradiation is characterized in Figure 6 by specimens 19-1 and 20-1, which represent an exposure of 1.45×10^{21} n/cm². Neither the yield strength nor the total elongation for these specimens varies greatly with direction, and only directional effects on fracture are preserved. That is, the transverse specimen undergoes an abrupt drop in load as an internal crack is initiated, and then the specimen shears to failure. The longitudinal specimen, however, necks slightly and breaks straight across by a noncleavage fracture.

A comparison of specimens 19-1 and 20-1 in Figure 6 (representing an exposure to 1.45×10^{21} n/cm² at 280 C) with specimens A7 and B7 in Figure 5 (representing an exposure to 2.3×10^{21} n/cm² at 60 C) reveals a strong temperature effect of irradiation. Not only do the specimens irradiated at the higher temperature show a greater hardening, but they also exhibit a marked departure in deformation behavior from their lower-exposure counterparts. In contrast, material irradiated at 60 C exhibits a single trend in basic plastic behavior and anisotropy over the entire exposure range.

3.3 Effects of Cold Work

The effects of cold work on the yield strength of Zircaloy-2 both before and after irradiation at 60 C are shown in Figure 7. In the unirradiated condition, cold rolling, aside from intensifying the crystallographic texture, increases the strength of Zircaloy-2 by about the same amount as does plastic strain from a uniaxial tensile test.

Other directional effects associated with strain hardenability in Figure 7 are also similar to the stress-strain curves plotted in Figure 2. A slight upward inflection in yield strength occurs beyond 40% cold work, which is attributed to the hardening contribution of new slip systems introduced at higher levels of cold work.

After irradiation to about 0.8×10^{19} n/cm², anisotropy in yield strength is relatively unchanged in annealed material but decreases in magnitude for the various cold work levels. Otherwise, the general features of the strength-versus-cold work relationships remain the same, including the inflection beyond 40% cold work. Directional relationships change noticeably with irradiation to higher exposure levels, however. Yield strength in the transverse direction increases with respect to the longitudinal direction in cold worked material and eventually surpasses it. This reversal in anisotropy occurs first at the higher cold work levels and then proceeds to occur at lower cold work levels with increased exposure. Although longitudinal yield strength for the cold worked conditions does not increase with exposure from 1.1×10^{20} to 22.7×10^{20} n/cm², a substantial increase occurs in the transverse direction. Consequently, yield strength is higher in the transverse direction for each cold work level at the highest neutron dose.

The curves in Figure 6 reveal two other interesting features: (1) yield strength anisotropy in annealed material is not greatly changed with irradiation, and (2) a slight reduction in longitudinal yield strength occurs for the cold work levels with an increase in neutron exposure from 1.1×10^{20} to 22.7×10^{20} n/cm². These results

indicate that a form of cold work recovery takes place at temperatures substantially below those required for rapid self diffusion. The difference in response to this recovery with test direction is similar to the differences in recovery shown in Figure 4.

The effects of irradiation at 60 C on the relationship between uniform strain and cold work are shown in Figure 8. In the unirradiated condition, uniform strain in both directions is greatly reduced by 10% cold work, but is slightly increased with higher amounts of cold work (refer also to Figure 2). In the longitudinal direction, uniform strain for annealed material is progressively reduced by neutron irradiation to approximately 3% for a dose of 22.7×10^{20} n/cm². In addition, the reduction in uniform strain with 10% cold work and the subsequent increase with higher amounts of cold work is preserved at each exposure. After irradiation to 22.7×10^{20} n/cm², substantial recovery in uniform strain occurs at cold work levels greater than 20%. Uniform strain at the 70% cold work level is nearly equal to the unirradiated value and slightly exceeds the value for annealed material at the same exposure.

Irradiation has a different effect on transverse uniform strain. After a nominal dose of 0.8×10^{19} n/cm², uniform elongation is lowest in the annealed condition and increases almost linearly with cold work. Uniform strain continues to decrease with continued irradiation (to a somewhat greater extent with higher cold work) until at 22.7×10^{20} n/cm², uniform strain is nearly independent of cold work, varying from 0.8 to 1.4% over the range 0 to 70% cold work.

From a designer's viewpoint, both the stress and strain at the maximum load point in a tensile test have significance, and both taken together define a point of instability beyond which continued elongation occurs with decreasing load. Figure 9 shows displacements in the point of plastic instability caused by both cold work and neutron irradiation at 60 C. At each neutron exposure level, the pair of curves representing the two principal working directions diverge at 0% cold work. The most unstable condition exists in annealed, transverse specimens, which exhibit nearly perfect plasticity; that is, necking sets in immediately after yielding, and the true stress-strain curve is approximated by a straight, horizontal line. Prior cold work has the benefit of increasing both the stress and strain at instability.

The effect of irradiation at 280 C on the relationship between yield strength and cold work is shown in Figure 10. Prior to irradiation, all specimens in the series were given a thermal treatment at 400 C for 48 hr to stabilize the cold worked structure with respect to the irradiation temperature. Because of differential recovery with test direction, longitudinal yield strength lies below transverse yield strength at each cold work level, in contrast to the cross over that occurs for as-rolled material (see Figure 7).

The effects of an elevated irradiation temperature can again be described by reference to three characteristic exposure zones: (1) a low-exposure zone, represented by curves 1, 2, 6, and 8, (2) a transition zone, represented by curves 5 and 7, and (3) a post-transition zone, represented by curves 11, 12, 19, and 20.

Irradiation to a nominal dose of 5×10^{19} n/cm² causes little change in the basic relationship between yield strength and cold work for both test directions, other than increasing the magnitude of yield strength about an equal amount for each cold work level and reducing anisotropy. Longitudinal yield strength is increased only slightly by higher doses to 1×10^{20} and 3×10^{20} n/cm², as shown by curves 6 and 8.

The appearance of curves 5 and 7, representing the transverse direction, differs markedly from that of curve 1, in that yield strength is about the same for annealed and 20% cold-worked material. The difference in yield strength between 20 and 40% cold-worked material differs with exposure, however. This difference is about the same for curves 1 and 5 (0.4×10^{20} and 1.0×10^{20} n/cm², respectively), but increases with higher doses.

Curves 19 and 11, representing neutron doses of 1.45 and 1.9×10^{21} n/cm², respectively, reveal a character similar to Curve 7; namely, little difference in transverse yield strength between 0 and 20% cold work, but a marked increase between 20 and 40% cold work. Curves 20 and 12 exhibit little effect of cold work on longitudinal yield strength. Unlike material irradiated at 60 C in the as-rolled condition, there are no signs of yield strength recovery due to irradiation at any of the cold work levels.

The effects of 280 C irradiation on the relationship between uniform strain and cold work are shown in Figure 11. Uniform strain in unirradiated, cold-worked material is substantially higher in the longitudinal than transverse direction because of differential recovery

from pre-irradiation annealing. Reductions in uniform strain with 280 C irradiation are similar in nature to those shown in Figure 8 for 60 C irradiation, and some cold work recovery effects are evident in the longitudinal direction. For example, curve 6 lies above curve 2 in spite of a higher neutron dose, and curve 8 has an inordinately high value for uniform strain in the 40% cold work condition. Curves 20 and 12, representing doses above about 5×10^{20} n/cm², show an independence of uniform strain with cold work. Transverse uniform strain falls to about 1% for all cold work levels at the lowest exposure, and undergoes further small reductions at higher exposures as yielding becomes more abrupt.

The effects of directionality on the room-temperature fracture behavior of cold-worked Zircaloy-2 irradiated at 280 C is illustrated in Figure 12. The longitudinal specimen undergoes almost a straight-across break and exhibits little or no shear lip. Conversely, the transverse specimen fractures almost entirely by shear. Disruptions in the oxide film of the specimens at shear zones give a graphical portrayal of the confinement of deformation during necking. During the necking of transverse specimens, plastic deformation occurs primarily along two macroscopic shear zones which tend to cross at the center of the specimen face. Necking continues until fracture occurs at the center of the specimen and spreads to zones of macroscopic shear; then, the crack spreads to the specimen surface along these zones.

Deformation in the neck of longitudinal specimens is less confined. The region of deformation extends further along the tensile

axis and develops observable shear zones only after the central crack approaches the surface. Lateral restraints imposed by such a neck develop a state of triaxial stress and favor fracture by combined tearing and shear.

The general fracture behavior illustrated in Figure 12 is common for all cold work levels, except that shear failure in the transverse direction is more extensive the higher the amount of cold work. Such a shear failure, when fully developed, is accompanied by small values of reduction of area and axial necking strain.

3.4 Elevated Temperature Tensile Behavior

Since most zirconium-alloy structural components are subjected to hot-water irradiation service, mechanical properties at 300 C are of practical interest. However, properties at this temperature are also of basic interest for two reasons: (1) deformation by mechanical twinning is less prominent at 300 C, whereas kink band development by basal slip becomes an important mode of deformation,⁽¹⁵⁾ and (2) radiation-induced defects generated at 280 C might affect deformation mechanisms differently at 300 C than at room temperature.

Relationships between 300 C yield strength and cold work for various neutron doses at 280 C are plotted in Figure 13. A comparison of these curves with those plotted in Figure 10 reveals some important differences. Yield strength anisotropy at 300 C in 0 and 20% cold-worked material is much less than that at room temperature for all neutron exposures, presumably because of a greater number of operative slip systems at the elevated temperature. However, 40% cold-worked material still exhibits a sizable anisotropy in 300 C yield strength

both before and after irradiation. Another difference is the absence at 300 C of the abnormally high transverse yield strength for annealed material observed at room temperature for curves 5 and 7, revealing a reduction in radiation-defect impedance to deformation. A similarity between room temperature and 300 C yield behavior is the small effect of cold work on longitudinal yield strength for doses of 5×10^{20} n/cm² and greater, as shown by curves 12 and 20.

Changes in the character of the stress-strain curves for transverse specimens at elevated temperature both before and after irradiation are illustrated in Figures 14 and 15. Figure 14, containing stress-strain curves for unirradiated material, shows that aside from a slight increase in uniform strain and a considerable decrease in flow stress at 300 C, the general appearance of the stress-strain curves for annealed Zircaloy-2 at both testing temperatures is similar. The curves for cold-worked material at the elevated temperature exhibit both a reduction in uniform elongation and a more-linear necking region of the curve. The latter effect is associated with a 45° shear failure. Both of these effects are caused by a marked reduction in strain hardenability with increased testing temperature and are somewhat more prevalent in 20 than in 40% cold-worked material.

The stress-strain curves in Figure 15 for material irradiated to a nominal dose of 5×10^{19} n/cm² show a rather marked reduction in uniform elongation. The room-temperature curves for cold-worked material again exhibit a linear necking region associated with the shear fracture illustrated in Figure 12. The elevated-temperature curves show a different behavior, however. Upon yielding, the cold

worked material undergoes an abrupt drop in flow stress during localized necking followed by a second, linear, precipitous drop in stress to failure. The form of these stress-strain curves is decidedly different, and signals a change in fracture mechanism. The nature of this new form of fracture is illustrated in Figure 16.

The stress-strain curve given in Figure 16 represents 20% cold-worked Zircaloy-2 irradiated to 5×10^{20} n/cm² at 280 C. The stress-strain curve exhibits a large yield drop at A, a smaller drop at B, a sharp drop at D, and final separation at E. The linear segments of the stress-strain curve connecting these points indicate that macroscopic shear processes are operating. The photographic inserts illustrate the region of necking at each of the critical points. The initial sharp yield point is associated with a shear zone established across the specimen face at 45° to the applied stress. At point B a conjugate shear zone is developed by bending moments in the load train imposed by the specimen offset. Both conjugate shear processes operate until a crack opens up in the center of the specimen (Point D). This crack grows by multiple shear as a diamond-shaped hole until internal and external surfaces meet at two junctures, and the sample separates at E as a symmetrical cup-cone failure. This form of failure is typical of a perfectly plastic material having no strain hardenability. Ductility was actually doubled by axial restraints from the testing method which initiated the second shear process. Otherwise, the specimen would have separated along a single shear zone. While failure along a single shear zone produces little reduction of area, failure involving conjugate shear zones produces considerable reduction of area. The

double cup-cone process did not operate at room temperature because of a much lower ratio of bending moment to shear stress.

3.5 Damage Accumulation Relationships

Yield strength values for annealed Zircaloy-2 irradiated at 280 C and tested at both room temperature and 300 C are plotted as a function of neutron flux ($E > 1$ Mev) in Figures 17 and 18. The curves for both testing temperatures are approximately parallel throughout the exposure range, and both exhibit a double inflection near 5×10^{20} n/cm². The difference in yield strength between 20 and 300 C is approximately 30,000 psi before irradiation, 32,000 psi with exposure to 4.5×10^{20} n/cm², and about 40,000 psi beyond 5×10^{20} n/cm².

Plots of yield strength as a function of log neutron exposure for both longitudinal and transverse specimens irradiated at both 60 and 280 C are presented in Figure 19. The exposure range is divided into three zones corresponding to the three characteristic types of yield behavior described earlier. Although straight-line functions have been fitted to the data for both temperatures of irradiation, the function is only well behaved at the lower temperature for the few points available. Nevertheless, a large difference in the accumulation of damage with irradiation temperature is quite clear, with the greater rate of hardening occurring at the higher temperature, contrary to normal expectation.

Back-extrapolation of the data for 280 C irradiation indicates a trend toward less damage at 280 C than at 60 C for exposures less than about 1×10^{20} n/cm² for the transverse direction and 5×10^{19} n/cm² for the longitudinal direction. The 280 C data in zone II are badly

scattered, with transverse data falling generally above the trend curve and longitudinal data falling below and not far from the 60 C curve. Within zone III both the transverse and longitudinal data follow extensions of the 280 C trend curves. An interpretation of these results would be that a marked transition in the rate of damage accumulation (based on a semi-log plot) takes place beyond 9×10^{19} n/cm² in the transverse direction and beyond 4.5×10^{20} n/cm² in the longitudinal direction. This shift in apparent transition with test direction suggests a directional response to irradiation damage in zone II apart from the otherwise invariant anisotropy with neutron dose.

Changes in yield strength caused by neutron damage are plotted against several functions of integrated neutron dose in Figure 20. Two of the functions are power laws with exponents of 1/2 and 1/3, and the third is an inverse exponential function with exponent 1/2 proposed by Makin and Minter.⁽²⁵⁾ Again considerable scatter exists in data for zone II, especially points 5, 7, 13, 14, 15, and 16. Discounting these points for the moment, all three functions fit the remaining data equally well to a dose of about 6×10^{20} n/cm², about as high as most correlations against a power law have been carried. A power law diverges more and more from the data at higher exposures and predicts infinite hardening at infinite exposure.

The inverse exponential function fits the data reasonably well throughout the exposure range, and can be graphically differentiated to determine a value for damage saturation. From the plot of Figure 20, the value of A (the change in yield strength at saturation) is 59,000

psi, which is larger than the initial yield strengths in both longitudinal (49,000 psi) and transverse (57,000 psi) directions.

The inverse exponential function,

$$\Delta \sigma_y = A \left[1 - \exp(-B\phi t) \right]^{1/2},$$

can be rearranged to give the following:

$$\ln \left[A^2 - \Delta \sigma_y^2 \right] = \ln A^2 - B\phi t.$$

A plot of $\ln \left[A^2 - \Delta \sigma_y^2 \right]$ against ϕt should yield a straight line with slope B (the rate of volume disturbance per unit volume). Such a plot is presented in Figure 21.

Referring to Figure 21, the envelope enclosing the points shows a variation in B from 0.58 to $2.9 \times 10^{-21} \text{ cm}^2/\text{n}$. The overall envelope can be divided into three subenvelopes representing three neutron flux ranges for the enclosed data points. A flux dependence in B is indicated. Such a dependency can be determined by plotting the slope of each point in Figure 21 as a function of flux. The results are given in Figure 22.

Although the data are badly scattered in Figure 22, there is a definite trend toward a decrease in B with increase in ϕ . From the interpretation of Makin and Minter,⁽²⁵⁾ B can be expressed as $B = \underline{a}\underline{v}$, where $\underline{a}$ is the number of obstacles generated per neutron, and $\underline{v}$ is the effective volume of each neutron-induced obstacle to deformation. It is not possible, from the data presented, to show whether it is a decrease in $\underline{a}$ or $\underline{v}$ which results in a decrease in the product $\underline{a}\underline{v}$ with neutron flux. Also, further study may reveal a time as well as a flux dependency in B, since agglomeration near the threshold of rapid self diffusion should be time dependent.

3.6 Tensile Ductility as a Function of Exposure and Hydrogen Content

From the foregoing descriptions of the plastic behavior of Zircaloy-2, it is evident that ductility as measured by the usual conventional parameters is markedly reduced by neutron irradiation both at 60 and 280 C. However, care must be taken in interpreting the nature of fracture from the magnitude of these parameters, since small values of reduction of area, uniform strain, or necking strain can be associated with extremes in either plasticity ^{or} ~~of~~ brittleness.

One of the uncontrolled variables in the 280 C irradiations was the pickup by the Zircaloy-2 of hydrogen generated by hot-water corrosion. Since the solubility of hydrogen in Zircaloy-2 is extremely low at room temperature, nearly all of the hydrogen absorbed is precipitated in the form of hydride platelets. The presence of such hydrides are known to cause embrittlement at room temperature, particularly if the hydrides are precipitated predominantly normal to the tensile axis. Studies by Parry and Evans⁽²⁶⁾ demonstrate that the tensile properties of hydrided Zircaloy-2 are restored in the range 100-200 C due to an apparent plasticity of the hydrides. Their data for the variation in ultimate tensile strength and reduction of area with temperature for Zircaloy-2 containing 0 and 200 ppm hydrogen are presented in Figures 23 and 24.

Two factors have an important bearing on the effects shown in Figures 23 and 24. First, the data represent short-transverse (through the thickness of the plate) specimens taken from cold-worked plate having an ultimate tensile strength exceeding 100,000 psi at room temperature. Secondly, both the basal planes and hydride platelets

were aligned normal to the tensile axis (the basal planes due to crystallographic texture and the hydride platelets due to precipitation alignment along planes of prior plastic flow). Longitudinal specimens having hydride platelets aligned parallel to the tensile axis exhibited little effect of 200 ppm hydrogen on room-temperature properties.

Hydrogen analyses of specimens irradiated in the present study showed a nearly linear increase in hydrogen content with time of hot-water exposure to a value of 175 ppm hydrogen for a dose of 1.5×10^{21} n/cm². The hydride platelets were aligned parallel to the rolling plane in cold-worked specimens, which placed the platelets parallel to the tensile axis for both longitudinal and transverse specimens. A similar hydride alignment existed in annealed material; however, a significant fraction of the hydrides were randomly oriented as well. From the results of Parry and Evans,⁽²⁶⁾ one would expect little effect of hydrides on room temperature properties for the above conditions. However, the structural damage caused by neutron irradiation is not equivalent to that caused by cold work, and the embrittling effects of hydrogen may be different in the two structures for the same strength level.

The effects of both 280 C irradiation and testing temperature on longitudinal necking strain are shown in Figure 25. The effect of test temperature is small throughout the exposure range. Furthermore, most of the ductility loss occurs at exposures where hydrogen is in solution at 300 C, indicating that most of the loss in ductility results from radiation damage. Radiation damage also caused a substantial loss in

necking strain for specimens irradiated at 60 C, for which hydrogen concentration remained below 15 ppm throughout their exposure. Necking strain for these specimens dropped from 16 and 22% for unirradiated longitudinal and transverse specimens, respectively, to approximately 5 and 3% after 2.2×10^{21} n/cm² exposure.

The effects of 280 C irradiation and testing temperature on reduction of area are illustrated in Figure 26. At room temperature, the transverse curve crosses the longitudinal curve at about 5×10^{20} n/cm², and falls to values less than 10% beyond 1.4×10^{21} n/cm² exposure. In contrast, reduction of area at 300 C is greater in the transverse direction throughout the exposure range.

Much of the above difference in directional effect with test temperature and neutron dose is related to differences in necking processes. At room temperature, necking in the transverse direction occurs primarily by shear along a single zone as illustrated in Figure 12. As irradiation proceeds and necking becomes more confined, reduction of area becomes less in the transverse direction, and a cross-over in directional relationships occurs. At 300 C, however, necking in transverse specimens occurs by the double cup-cone mode shown in Figure 16, and transverse reduction of area remains greater throughout the exposure range.

For equal fractions of hydrides oriented normal to the test direction, hydrogen embrittlement is probably more severe in the transverse than longitudinal direction because of a greater restriction in slip systems, the occurrence of twinning, and a greater confinement of deformation during necking. The abrupt drop in transverse reduction

of area at 300 C for a dose of 2×10^{21} n/cm² signals the advent of hydrogen embrittlement at this temperature. The convergence with irradiation of longitudinal reduction of area at room temperature and 300 C indicates that radiation damage probably has an overriding effect on ductility loss in this direction throughout most of the exposure range.

3.7 Post-Irradiation Recovery

The recovery of irradiation damage in Zircaloy-2 has been studied by Howe⁽²⁷⁾ and by Howe and Thomas⁽²⁸⁾ on material irradiated both at 50 and 280 C. Isochronal annealing curves for samples taken from the axial direction of pressure tubing and annealed prior to irradiation are plotted in Figure 27. In general, material studied by Howe and Thomas is similar in crystallographic texture and tensile properties to longitudinal specimens taken from rolled plate in the present study. Allowing for differences in reporting neutron flux (with respect to energy cutoff), the as-irradiated values for yield strength in Figure 27 for both 50 C and 280 C irradiations compare closely with corresponding longitudinal values in Figure 19. Furthermore, similar effects of irradiation temperature on neutron damage are established by both studies.

Referring to Figure 27, material irradiated at 280 C to 7.7×10^{19} n/cm² requires a slightly higher recovery temperature than material irradiated at 50 C to a nearly comparable exposure. Howe⁽²⁷⁾ also found for the material irradiated at 280 C a slightly higher activation energy (2.2 ev versus 1.8 ev) and a slight difference in reaction order, indicating that the nature of the damage is somewhat different.

Higher neutron doses at 280 C increase the recovery temperature much further, revealing an increased complexity in the damage.

Annealing treatments at 360 C for 10 hours were conducted on specimens representing all levels of cold work and neutron exposure (greater than 1×10^{20} n/cm²) in the present study. Aside from only a few exceptions (about 5 to 15% recovery), no observable recovery occurred. These results indicate a shift in recovery to still higher temperatures than those shown in Figure 27. Such an increase in damage stability might result from small differences in temperature and flux-rate near 300 C. Although the inlet loop water in the present study was controlled at $280 \text{ C} \pm 2 \text{ C}$, the sensible specimen temperatures measured by thermocouples varied from 288 to 304 C. Even these slightly higher irradiation temperatures near the threshold for rapid self diffusion might cause a greater thermal stability of the defected structure. Furthermore, the scatter in yield strength data in zone II of Figure 19 and the apparent flux rate dependency shown in Figure 22 indicate that details associated with flux might be important.

4.0 DISCUSSION

The nature of irradiation damage in Zircaloy-2 and the influence of this damage on deformation and fracture mechanisms are discussed below in the light of present knowledge for hcp metals and alloys. Unfortunately, very few basic studies have been conducted on irradiated, hcp metals; and basic mechanisms for the unirradiated state are not completely understood. However, arguments are presented as a basis for further investigation.

4.1 Nature of Radiation Damage

There is growing evidence that two general types of defects are effective in irradiation hardening; namely, vacancy clusters and interstitial loops. Vacancy clusters are considered to form directly from displacement spikes of primary knock-on atoms;^(29,30) whereas, interstitial loops are nucleated at lattice regions of momentary interstitial supersaturation. Nucleation is held to be heterogeneous,⁽³⁰⁾ although nucleation mechanisms have not been definitely established. Interstitial defects must diffuse to nuclei if loops are to achieve a size visible in the electron microscope ($> 50 \text{ \AA}$). At the lowest temperature where interstitial loops can grow, vacancies are virtually immobile in the lattice.⁽²⁹⁾

Although clustered defects in the form of spots and dislocation loops have been observed in irradiated bcc^(29,31) and fcc^(30,32) metals, only a few observations have been reported for irradiated hcp metals. Price⁽³³⁾ observed vacancy-type dislocation loops in cadmium subjected to ion bombardment at -90 C . Roy⁽³⁴⁾ observed black dots, varying in size from ~ 65 to $4000 \text{ \AA}$, in foils of zirconium irradiated to $2.7 \times 10^{18} \text{ n/cm}^2$ at 40 C . These defects gave contrast effects similar to dislocation loops on $\{10\bar{1}0\}$ planes, but a positive identification was not made. Walters, Van der Walt, and Makin⁽³⁵⁾ observed prismatic loops lying principally on $\{11\bar{2}0\}$ planes and in smaller numbers on $\{10\bar{1}0\}$, $\{1\bar{1}01\}$ and $\{11\bar{2}2\}$ planes in beryllium irradiated to $2 \times 10^{20} \text{ n/cm}^2$ at 350 C . Loops lying on $\{10\bar{1}0\}$ and $\{11\bar{2}0\}$ planes had $1/3 \langle 11\bar{2}0 \rangle$ Burgers vectors and did not exhibit stacking faults. Small defects ($\sim 35 \text{ \AA}$), presumably interstitial clusters or unresolvable interstitial

loops, have been observed recently by Brimhall and Mastel⁽³⁶⁾ in rhenium irradiated to an exposure of 7×10^{18} n/cm² at 40 C. Foils from this latter study exhibited confined channels of moving dislocations similar to those observed in molybdenum,⁽³⁷⁾ iron,⁽³⁸⁾ and copper⁽³⁹⁾ strained after irradiation.

Studies conducted on irradiated copper and nickel by Greenfield and Wilsdorf^(40,41) and on molybdenum by Meakin and Greenfield⁽²⁹⁾ show that irradiation temperature has a major influence on the nature of the resulting damage. In general, irradiations carried out at homologous temperatures (T/T_m) below about 0.26 do not cause visible damage, except at high doses.^(36,37) At homologous temperatures between about 0.27 and 0.40, large prismatic dislocation loops are observed after doses of about 10^{18} n/cm².⁽²⁹⁾ For zirconium, a homologous temperature of 0.27 corresponds to 300 C.

The observation that dislocation loops form predominantly along basal planes in cadmium compared to prismatic planes in beryllium is in agreement with the interplanar spacings for these metals. Whereas the c/a ratio for cadmium is greater than ideal and maximum interplanar spacing occurs between basal planes, the c/a ratio for beryllium is much less than ideal and maximum interplanar spacing occurs between prism planes. Further observations for other hexagonal metals are required to establish such a relationship, however, since electronic properties and Peierl's forces may also play an important role.

At ambient irradiation temperatures ($T/T_m \simeq 0.16$) it is expected that a large concentration of interstitial clusters will precipitate as stable loops in zirconium and Zircaloy and these will grow slowly

due to the large number of loops available for migrating interstitials. At elevated temperatures, however, the loop concentration will be much smaller due to the difficulty in achieving localized interstitial saturation and the larger loop size required for stabilization. However, the growth of stable loops would be rapid due to the reduced number of loops and the enhanced mobility of interstitials. The interactions among loops during rapid growth at these temperatures could result in complex dislocation networks similar to those observed by Price⁽⁴²⁾ in cadmium subjected to ion bombardment. These networks would differ from those formed by plastic strain due to the possible presence of stacking faults within the interacting loops.

The above model for interstitial loop formation and growth provides insight into the temperature-enhanced damage observed for Zircaloy-2. At ambient irradiation temperature, hardening results primarily from vacancy clusters and perhaps unresolvable interstitial loops, both of which providing pinning sites for existing dislocations. The initial rate of damage accumulation is reduced by raising the irradiation temperature because of the increased rate of defect recombination, the greater difficulty in nucleating stable dislocation loops, and the larger interobstacle spacing. The exposures involved for this behavior correspond to zone I in the present study. At higher exposures, however, a sufficient number of stable loops nucleate to create dislocation tangles as they grow and interact. These tangles add to the existing substructure and cause a marked increase in strength by a form of "internal cold working." The advent of this form of damage occurs in zone II or the transition zone. At still higher exposures

(zone III), the substructure continues to become more complex and radiation-hardening continues to increase, failing to saturate at the highest exposure achieved (2×10^{21} n/cm²).

Three observations from the data presented for Zircaloy-2 support the above argument. The first is the marked difference in the effect of cold work on yield strength between zones I and III in Figures 10 and 13. For pre-transition exposures, the relationship between yield strength and cold work is relatively unaffected, and neutron damage contributes a hardening which is additive to that caused by prior cold work. For post-transition exposures, however, longitudinal yield strength is nearly independent of cold work. A similar effect is also observed for transverse yield strength, except that a marked increase still occurs between 20 and 40% cold work which will be discussed later.

The second observation is the marked shift in post-irradiation recovery temperature shown in Figure 27 with an increase in neutron dose from 7.7×10^{19} n/cm² (zone I) to 2.7×10^{20} n/cm² (zone II or III). After high doses, the neutron damage recovery temperature is increased to temperatures where from Figure 4 significant cold work recovery is known to occur.

The third observation is the occurrence of a radiation-hardening transition in transverse yield strength for annealed material at exposures lower than required for longitudinal yield strength. Added structural impedance from dislocation tangles should have a stronger effect on transverse yield strength because of the small number of

potential slip systems and the need for stress concentrations to initiate twinning.

4.2 Yield Behavior and Anisotropy of Flow and Fracture

Observations of a yield point in irradiated hcp metals and alloys vary. Kemper and Zimmerman⁽⁴³⁾ reported a yield point in annealed Zircaloy-2 irradiated at 40 C; however, the yield point was removed by post-irradiation annealing at 300 C. Howe and Thomas⁽²⁸⁾ observed a yield point in Zircaloy-2 irradiated at 280 C during testing at 280 C, but not at room temperature. Similarly, Makin and Minter⁽⁴⁴⁾ observed a yield point in titanium irradiated at 100 C during testing at 200 C, but not at lower temperatures to -195 C. A yield point was only rarely observed by Bruch, et al,⁽⁴⁵⁾ in zirconium irradiated below 100 C and tested at room temperature. In the same study, a yield point was noted in titanium, but not until 1.5×10^{20} n/cm² exposure. One can conclude from these observations that while a yield point can develop in hcp metals, it is more likely to occur at elevated temperatures.

Yield point behavior in irradiated Zircaloy-2 from the present study is in general agreement with the above observations and shows directional effects similar to unirradiated material. Yielding in the longitudinal direction was continuous at room temperature throughout the exposure range, and a yield point rarely occurred at 300 C except after high neutron doses (greater than 1×10^{21} n/cm²). An abrupt yield point in transverse specimens normally occurred at 300 C and frequently occurred at room temperature throughout the exposure range. Room temperature yield points for transverse specimens irradiated at 50 C were more abrupt and required less pre-yield strain than samples

irradiated at 280 C. Transverse samples containing cold work also exhibited abrupt yielding at both room temperature and 300 C after exposures exceeding 5×10^{20} n/cm².

The above yield behavior is attributed to the sweeping out of defect channels by mobilized dislocations. (36,37,38,39,46) By absorbing defects, these dislocations reduce the stress required to move other dislocations in their wake. This is truly an unstable effect, since the stress required to initiate plastic flow is greater than the stress required to sustain plastic flow. This type of deformation causes coarse slip (extensive slip on well-spaced slip bands), (39) and the ability of the material to strain harden depends upon the number of slip systems participating in the deformation.

The strain hardenability of irradiated Zircaloy-2 is strongly influenced by the direction of applied stress. A Lüders band generally develops at 45° to the applied stress in transverse specimens, and plastic deformation is confined to this band until failure occurs. Coarse slip bands also form in longitudinal specimens; however, slip occurs along a large number of such bands throughout the gage section of the specimen. These strong directional effects in both the yield behavior and strain hardenability of irradiated Zircaloy-2 suggest differences in the interactions between dislocations and radiation-induced defects.

The interaction between edge dislocations in a prismatic slip plane moving with respect to screw dislocations in a conjugate prism plane creates jogs lying in the basal plane. Since the resolved shear stress for basal slip is very small in the longitudinal direction of

textured sheet, such jogs can be an effective drag to the motion of edge dislocations. If dislocation loops form on prism planes during neutron irradiation, the amount of such jogging increases rapidly with strain and causes significant hardening. Such interactions also form sessile jogs on screw dislocations which either lock them in place or promote extended loops emanating from jog-anchored dipoles.⁽⁴²⁾ As a consequence of these interactions, longitudinal specimens retain some ability to strain harden after irradiation.

Martin and Reed-Hill⁽¹⁵⁾ have found that basal slip can occur in transverse specimens of zirconium having a rolling texture. Furthermore, the cross slip of screw dislocations from prism to basal planes should be favored by the high stacking fault energy of zirconium. Such cross slip in zirconium and Zircaloy-2 has actually been observed by transmission electron microscopy.^(47,48) Although extended dislocations enclosing a stacking fault ribbon have also been observed,⁽³⁴⁾ these occurred in cold worked material where high flow stresses were required for dissociation. Dislocations on basal planes have the unique advantage of being unaffected by intersections with dislocations on prism planes. Since basal planes contain both basal and prismatic slip vectors, such intersections create jogs lying in the basal plane for both edge and screw components, which is a trivial case.

Upon yielding, therefore, relatively easy glide by basal slip can sustain deformation in the transverse direction until failure occurs. Since cross-slip is thermally activated, the process should be both temperature and rate sensitive. The abrupt yielding and

extreme plasticity exhibited by transverse specimens at 300 C reveal such a strong temperature effect on flow and fracture behavior.

4.3 Cold Work Effects

The extreme plastic instability of irradiated, transverse specimens in the pre-annealed condition results from large differences in the stresses required to initiate and sustain plastic flow. If little dislocation interaction takes place after channels through defects have been cleared, deformation remains localized and the material exhibits nearly perfect plasticity. Prior cold work has the benefit of adding impedance to dislocation motion and reducing the stress decrement between initiation and continuation of plastic flow. A sufficiently high flow stress can also dissociate dislocations into widely separated partials, inhibiting cross slip into basal planes.

Further reductions in the heterogeneity of deformation can be achieved by increasing the number of active deformation systems. For example, Reed-Hill⁽¹⁴⁾ has demonstrated that the room-temperature, transverse uniform strain of zirconium is substantially increased by a low-temperature, pre-straining operation which initiates most of the possible twinning systems. Similar pre-treatments could be beneficial in improving the uniform strain of irradiated material.

The effects of cold work on the properties of irradiated Zircaloy-2 vary with the amount of cold work, amount of exposure, temperature of irradiation, and direction of applied stress. The relatively small difference in the yield strength-versus-cold work relationship after all exposures at 60 C (Figure 7) and after zone I exposures at 280 C (Figure 10) indicate that radiation defects, aside from raising the

flow stress, do not override the hardening effect of strain-induced obstacles. The same cannot be said for zone II or III exposures at 280 C. The damage to transverse specimens at these exposures clearly overrides the effect of strain hardening to the 20% cold work level. At exposures beyond about 10^{21} n/cm², yield strength is nearly independent of cold work in the longitudinal direction. This behavior indicates that the structural damage done at these exposures is more complex than isolated dislocation loops or defect clusters and adds weight to the argument that loops grow and interact to form complex subnetworks at these exposures.

The nature of dislocation arrangements and interactions in cold-worked zirconium and Zircaloy-2 has been investigated by transmission electron microscopy. (34,47,48) Both zirconium and Zircaloy-2 exhibit similar features; namely, an absence of tangling for less than 15% cold work, relatively straight dislocation alignment, and an abundance of edge dislocation pile-ups at grain boundaries. At low strains, dislocations are confined primarily to $(10\bar{1}0)$ planes and interact primarily by torques induced by elastic stresses at the projected crossings of dislocation traces on parallel slip planes. (34,47)

Two characteristics of unirradiated Zircaloy-2 in the present study indicate that the nature of dislocation interactions changes at about 20% cold work: (1) the minimum in uniform elongation at about 20% cold work (Figures 8 and 9), and (2) the inflection in yield strength versus cold work beyond about 40% cold work (Figures 4 and 7). Taken together, these trends indicate that additional slip systems operate beyond about 20% cold work and contribute to both strain

hardenability and flow stress by the formation of complex dislocation tangles or networks.

The marked increase in yield strength in the transverse direction between 20 and 40% cold work after relatively high exposures at 280 C is attributed to the superposition of the substructure formed by loop interaction during irradiation upon the pre-irradiation dislocation structure. The effect of this highly complex structure on yield strength is again greater in the transverse direction because of texture restraints on deformation.

Cold work recovery effects during irradiation at 60 C (substantially below the temperature range for rapid self diffusion) have been observed not only for Zircaloy-2 in this study, but also for cold worked zirconium.⁽⁴⁴⁾ Similar processes in zinc⁽⁴⁹⁾ and cadmium,⁽⁵⁰⁾ involving mass transfer at temperatures too low for bulk diffusion, have been attributed to climb by pipe diffusion. This mechanism involves the preferential diffusion of vacancies along dislocation channels, and is favored if the sum of activation energies for vacancy formation and vacancy movement along dislocations is appreciably smaller than the activation energy for bulk diffusion. These conditions are best satisfied the larger the Burgers vector; therefore, pipe diffusion in Zircaloy-2 would be most favorable for dislocations on pyramidal planes. If it is assumed that pyramidal slip becomes progressively more active with increased cold work (probably accounting for increased strain hardenability beyond 20% cold work), an explanation is available for the increased recovery in uniform

elongation with cold work level exhibited by both curve G in Figure 8 and curve 8 in Figure 11.

4.4 Concluding Remarks

Although descriptions of irradiation effects to hcp metals and alloys have been largely based on zirconium and Zircaloy-2, attempts have been made to discuss these effects with reference to basic mechanisms applicable to a variety of hcp metals and alloys. Additional investigations are required to confirm the arguments proposed and to adequately define the morphology of radiation defects at various irradiation temperatures and neutron doses. However, it seems clear from available evidence that crystallographic anisotropy in plastic flow and fracture behavior is preserved and, in some cases, intensified after neutron irradiation. It follows, therefore, that crystal structure is a principal variable in irradiation damage to metals.

Within the hcp family of metals (for equal homologous temperatures), large differences in radiation effects are expected among the subgroups cadmium and zinc (c/a greater than ideal); magnesium (c/a near ideal); and zirconium, titanium, and beryllium (c/a less than ideal). Differences should arise not only in the crystallographic nature of the defects generated, but also in the interactions between these defects and the deformation systems characteristic of these subgroups. Such differences plus effects of crystallographic texture, which are relatively strong for hcp metals and alloys, will reveal unique damage effects not to be found in bcc and fcc metals. Future investigators should take advantage of these discriminating features to advance the basic understanding of damage mechanisms.

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FIGURE 1

POLE FIGURES FOR THE PRINCIPAL LATTICE PLANES IN ANNEALED AND 70% COLD-WORKED ZIRCALOY-2

TRUE STRESS-STRAIN CURVES TO THE LIMIT OF UNIFORM STRAIN SHOWING EFFECTS OF COLD WORK AND TESTING DIRECTION

FIGURE 3
DEFORMATION APPEARANCE IN LONGITUDINAL
(ABOVE) AND TRANSVERSE (BELOW) DIRECTIONS
(500X, B ETCH, POLARIZED LIGHT)

Longitudinal

Transverse

FIGURE 4
EFFECTS OF RECOVERY ANNEALING ON YIELD STRENGTH AND
UNIFORM ELONGATION OF ZIRCALOY-2

FIGURE 5
CONVENTIONAL STRESS-STRAIN CURVES FOR ANNEALED ZIRCALOY-2 TESTED IN TENSION AT ROOM TEMPERATURE BEFORE AND AFTER NEUTRON IRRADIATION AT 60 °C

FIGURE 6
CONVENTIONAL STRESS-STRAIN CURVES FOR ANNEALED ZIRCALOY-2 TESTED IN TENSION AT ROOM TEMPERATURE BEFORE AND AFTER NEUTRON IRRADIATION AT 280 °C

FIGURE 7

THE EFFECTS OF COLD WORK ON THE ROOM TEMPERATURE
YIELD STRENGTH OF ZIRCALOY-2 BOTH BEFORE AND
AFTER IRRADIATION AT 60 °C

FIGURE 8

THE EFFECTS OF COLD WORK ON THE ROOM TEMPERATURE
UNIFORM STRAIN OF ZIRCALOY-2 BOTH BEFORE AND
AFTER IRRADIATION AT 60 °C

FIGURE 9

DISPLACEMENT IN THE POINT OF PLASTIC INSTABILITY WITH COLD WORK
AND IRRADIATION

FIGURE 10
THE EFFECTS OF COLD WORK ON THE ROOM
TEMPERATURE YIELD STRENGTH OF ZIRCALOY-2
BOTH BEFORE AND AFTER IRRADIATION AT 280 °C

FIGURE 11
THE EFFECTS OF COLD WORK ON THE ROOM TEMPERATURE
UNIFORM STRAIN OF ZIRCALOY-2 BOTH BEFORE AND
AFTER IRRADIATION AT 280 °C

FIGURE 12

ROOM TEMPERATURE TENSILE FRACTURE APPEARANCE
OF ZIRCALOY-2 AFTER IRRADIATION (10X)

Oblique

Transverse

Profile

20% Cold Work
 6.4×10^{20} Neutrons/cm²
($E > 1$ Mev)

Oblique

Longitudinal

Profile

FIGURE 13
THE EFFECTS OF COLD WORK ON THE 300 °C YIELD
STRENGTH OF ZIRCALOY-2 BOTH BEFORE AND AFTER
IRRADIATION AT 280 °C

FIGURE 14
TRANSVERSE STRESS-STRAIN BEHAVIOR AT ROOM
TEMPERATURE AND 300 °C OF UNIRRADIATED
ZIRCALOY-2

FIGURE 15
TRANSVERSE STRESS-STRAIN BEHAVIOR AT ROOM
TEMPERATURE AND 300 °C OF IRRADIATED
ZIRCALOY-2 (4×10^{19} n/cm² at 280 °C)

FIGURE 16
TRANSVERSE FRACTURE BEHAVIOR AT 300 °C AFTER IRRADIATION
(5×10^{20} n/cm² at 280 °C)

FIGURE 17
LONGITUDINAL AND TRANSVERSE YIELD STRENGTH AT ROOM
TEMPERATURE AS A FUNCTION OF NEUTRON DOSE AT 280 °C

FIGURE 18
TEMPERATURE DEPENDENCE OF LONGITUDINAL YIELD STRENGTH
FOR ANNEALED ZIRCALOY-2 AS A FUNCTION OF NEUTRON DOSE
AT 280 °C

FIGURE 19

YIELD STRENGTH OF ANNEALED ZIRCALOY-2 ASA FUNCTION OF NEUTRON EXPOSURE AT VARIOUS TEMPERATURES

FIGURE 20

FUNCTIONAL RELATIONSHIPS BETWEEN YIELD STRENGTH AND NEUTRON DOSE

FIGURE 21
 LINEAR REPRESENTATION BETWEEN YIELD STRENGTH AND NEUTRON
 DOSE FOR THE INVERSE EXPONENTIAL RELATIONSHIP

FIGURE 22
 VARIATION IN COEFFICIENT B FROM THE INVERSE EXPONENTIAL
 RELATIONSHIP WITH NEUTRON FLUX

FIGURE 23

ULTIMATE TENSILE STRENGTH FOR COLD-WORKED
ZIRCALOY-2 CONTAINING HYDRIDES PRECIPITATED
NORMAL TO TENSILE AXIS

FIGURE 24

REDUCTION OF AREA FOR COLD WORKED ZIRCALOY-2
CONTAINING HYDRIDES NORMAL TO TENSILE AXIS

FIGURE 25
EFFECT OF NEUTRON IRRADIATION AND TESTING TEMPERATURE
ON LONGITUDINAL NECKING STRAIN

FIGURE 26
EFFECT OF NEUTRON IRRADIATION AND TESTING TEMPERATURE
ON PERCENT REDUCTION OF AREA IN BOTH THE LONGITUDINAL
AND TRANSVERSE DIRECTIONS

FIGURE 27

ISOCHRONAL RECOVERY CURVES FOR ANNEALED ZIRCALOY-2
IRRADIATED AT BOTH 60 °C AND 280 °C

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