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NEON ION SIMULATION OF NEUTRON INDUCED IRRADIATION
GROWTH IN ZIRCONIUM ALLOYS

by .

Rodney Herring

A Thesis
Submitted to the Faculty of Graduate Studies and Research
Through the Department of Engineering Materials
in Partial Fulfillment of the Requirements for
the Degree of Master of Applied Science
at the University of Windsor

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ABSTRACT

Irradiation growth, a shape change in the absence of an applied stress, is one of the factors limiting the performance of pressure tubes in CANDU-PHW reactors. Experimental data on irradiation growth in zirconium and its alloys that already exists in the literature is critically reviewed, as well as, the present models proposed for irradiation growth. New data is presented for growth in zirconium, zircaloy-2, Zr-2.5wt%Nb and the XL Alloy in which neon ion bombardment is used to simulate the reactor environment. The irradiation conditions and effects of microstructure are considered in full and recommendations are made with respect to future growth experiments.

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CHAPTER 1

1.1 Introduction

The dimensional changes of pressure tube material in CANDU-PHW* power reactors consist of two components, irradiation induced growth and irradiation enhanced creep. Creep results from the application of an externally applied stress, whereas, irradiation growth is a process which in the absence of any applied stress results in a macroscopic shape change of a material during irradiation with little or no volume change. CANDU pressure tubes manufactured from zirconium-based alloys are 6 metres in length and can elongate up to 5 cm during their design lifetime due to irradiation growth and creep, putting significant design limitations and setting life expectancies of about 30 years on the reactor's structural material. These limitations can be better designed to if there is a better understanding of the factors contributing to irradiation growth.

The growth phenomena was first reported at the Second Conference on Peaceful Uses of Atomic Energy where it was shown that a most spectacular effect of irradiation upon uranium metal is manifested by radical changes in dimensions (1). Orthorombic alpha uranium elongated in the $[010]$ direction, shortened in the $[100]$ direction and remained approximately unchanged in the $[001]$ direction. The existence of irradiation growth in zirconium was first established by Buckley (2). He measured the linear expansion and change in

curvature of bent thin foils of annealed zirconium irradiated with fission fragments. Recently, a similar technique using ion simulation of neutron irradiation damage has been used to measure the irradiation growth and creep rates of polycrystalline zirconium and its alloys (3,4).

The theory of growth proposed by Buckley (2) was applied to many metals. The principal requirement of the theory is that the metal should have both crystallographic and thermal expansion anisotropy. An incoming neutron that initiates an energetic primary knock-on collision creates a cascade of displaced atoms (the displacement spike) and a high localized generation of heat (the thermal spike). In Zirconium the thermal expansion coefficient is greatest along the c-axis direction, so the cascade produced during irradiation will experience the greatest compressive stresses along this direction. This favours the collapse of vacancy clusters into dislocation loops having a Burger's vector parallel to the c-axis. The interstitials produced during irradiation were believed to condense as loops with a-type Burger's vectors, i.e., $a/3\langle 11\bar{2}0 \rangle$. This mechanism is equivalent to a transfer of atoms from the basal plane to the prism plane (5). Later, a TEM study (6) of uranium supported Buckley's theory. A study of zirconium irradiated by 150 KeV Krypton ions to a dose of 10^{17} ions/m² revealed dislocation loops with $a/3\langle 11\bar{2}0 \rangle$ Burger's vectors, distributed evenly on all three possible $\{10\bar{1}0\}$ planes, but no c-type dislocations were reported (1).

For many years Buckley's theory was accepted for

zirconium, until in 1969 it was questioned by Kelly and Buckley (8). They found no apparent correlation between measured and expected (from Buckley's model) growth of neutron irradiated specimen of Zircaloy-2. Later, Northwood et al (9) in a 'round-robin' experiment confirmed that no c-type dislocations are formed for annealed Zircaloy-2 irradiated to 10^{25} n/m² ($E > 1$ MeV) and that only dislocation loops with $a/3 \langle 11\bar{2}0 \rangle$ Burger's vectors were formed, comprising a mixture of vacancy and interstitial loops.

Recently, irradiation growth results by Adamson, Tucker and Fidleris (10) along with TEM analysis of the same specimen by Gilbert and Holt (11), revealed the many important factors affecting growth and suggested other possible mechanisms for the growth process.

The factors affecting the growth process have been shown to include the operating conditions (neutron fluence and temperature); microstructure (dislocation density, grain size and shape); crystallographic texture (anisotropy); and fabrication procedure (residual stresses) (12).

In this thesis, a critical review of the available growth data will be presented, together with a detailed discussion of the roles of the various factors affecting growth. As will become evident in this review, much of the delay in determining the role of the various factors affecting growth is due to the long length of time needed to neutron irradiate test specimens to a high dose. With these limitations in mind an experimental technique originally

developed by Parsons and Hoelke (3,4), where neon ions simulate neutron irradiation growth and creep on cantilever beam type specimens, has been used to generate information on growth of zirconium alloys in a relatively short period of time. It has been shown that such an experimental technique has considerable promise in studying this very important technological area. In addition to the basic neon ion irradiations and growth measurements, electron microscope techniques have been used to study the nature and character of the irradiation damage giving rise to the measured irradiation growth.

CHAPTER 2

2.1 Critical Review of Data on Irradiation Growth in Zirconium Alloys

This review is divided into a number of sections and subsections. The early sections contain growth data for single crystals, annealed polycrystals and cold-worked materials. The next sections deal with the effects of irradiation variables such as flux (fluence) and temperature, then material variables (e.g., microstructure, texture and composition). These are followed by short sections on post-irradiation annealing effects and the conservation (or not) of volume. Detailed consideration is then given to the proposed models for irradiation growth. The review concludes with a discussion of the experimental methods used to measure irradiation growth.

2.1.1 Single Crystals

Buckley (23) in 1962, was the first to report data for single crystals and he found an expansion in the direction of the a-axis and a contraction along the c-axis, with no overall volume change. However, Kelly (13) in reporting data of Hilditch for a spherical single crystal irradiated to a fluence of $7 \times 10^{24} \text{ n/m}^2$ also noted an a-axis expansion but a slight expansion in the c-axis direction. Northwood et al (14) for a single crystal rod orientated close to the c-axis showed an initial c-axis expansion of 5×10^{-4} which reversed after a fluence of about $6 \times 10^{23} \text{ n/m}^2$ so that ultimately a length decrease occurred.

The latest data by Carpenter et al (15), on specimens orientated close to either the a-axis, the (a+c)-axis or the c-axis during irradiation are shown in Figure 1. At 353K, the specimens expanded in the a-axis direction with a saturation strain of around 1.6×10^{-4} , contracted in the c-axis direction with a saturation strain of $\sim 1 \times 10^{-4}$ and very slightly contracted in the (a+c)-axis direction. Further, for single crystal specimens irradiated at 553K there was an expansion in the a-axis direction, which saturated at $\sim 1.4 \times 10^{-4}$ and contraction in the c-axis direction, which saturated at $\sim 0.8 \times 10^{-4}$, (i.e., both slightly less than for the specimens irradiated at 353K). Zone refined and iodide purity specimens irradiated at 553K are compared in Figure 2 to show the effect of purity on growth. It can be seen that the less pure specimens (zone refined) saturate much faster in the a-axis direction, whereas the two types of specimens contract at an almost equal rate in the c-axis direction. The saturation growth strains being equal in both a- and c-axis directions. Carpenter et al (15) considered the various growth mechanisms responsible for the a-axis expansion and c-axis contraction. The a-axis expansion was thought to be due to a net annihilation of interstitials at a -type dislocations and interstitial loops. They were unsure of the cause of the c-axis contraction but suggested three possible mechanisms: i) elastic relaxation around vacancies or small vacancy clusters; ii) sub-microscopic vacancy loops with c component Burger's

vectors, (undetectable by TEM) or iii) a non-Hookian contraction around small dislocation loops on prism planes due to dislocation core effects. Carpenter et al (15) then demonstrated that lattice relaxation around vacancies could not account for the c-axis contraction although some contribution is possible. Secondly small $\langle c \rangle$ -component loops are unlikely for two reasons: a) since $\langle c \rangle$ -component dislocations are essentially the same mechanism for contraction as that for uranium, it would be fortuitous if the growth of zirconium single crystals would saturate so rapidly and completely while the opposite behaviour occurs in uranium, b) if it is necessary to assume that the $\langle c \rangle$ -component loops are relatively unstable, their high energy will tend to mitigate against their formation in cascades. Thirdly, non-Hookian behaviour at the cores of the $\langle a \rangle$ -type dislocation loops would require a very high dislocation density $\sim 2 \times 10^{23} \text{ m}^{-3}$ at 2nm in diameter, based on Goland and Keating's (16) calculation. This mechanism may be reasonable in view of the low irradiation temperatures used.

2.1.2 Annealed Polycrystalline Material

The data for polycrystalline zirconium, Zircaloy-2 and Zircaloy-4 are summarized in Figure 3, with additional information provided by the key in Table 1, which lists for each symbol used in the figure, the irradiation temperature, texture coefficient, type of material and the reference. The texture coefficient of a polycrystalline material is the state of distribution of crystal orientations. The growth strains versus neutron fluence are plotted on a log-log scale.

The growth of polycrystalline zirconium and its alloys exhibit an initial rapid transient, followed by a saturating growth strain which is about an order of magnitude greater than single crystal saturation. The initial transient in growth can be either positive or negative according to Fidleris (12), which he attributes to the recovery of residual stresses.

If the empirical relationship between the growth strain, ϵ , and the neutron fluence (ϕt) is given by:

$$\epsilon = \text{constant} \times (\phi t)^q \quad (2.1)$$

then there are essentially two regions for the growth strain versus fluence curves. The initial growth rate is high and for fluences $< 6 \times 10^{24} \text{ n/m}^2$, the best fit q value is 0.65. Above this value, the best fit q value is 0.11. There are two exceptions to the data. Adamson's data at 673K and 653K, the highest measured irradiation temperatures, which show no tendency towards saturation of growth, having q values of 0.69 and 0.43, respectfully.

The growth saturation in annealed polycrystalline material, as well as, single crystals is taken as evidence that point defect annihilation has virtually reached a steady state, such that equal fluxes of interstitials and vacancies are lost to each type of sink, giving zero strain rate and/or recombination is very high.

Fidleris (21) advises not to use equation (2.1) because he has shown that early transients affect the slope of the logarithmic plot.

2.1.3 Cold Worked Material

The data for cold-worked materials include both cold-worked material and material cold-worked plus stress relieved which is the common condition for pressure tubes in both CANDU* and SGHWR⁺, and fuel cladding in BWR^x and PWR[†] reactors.

The main differences in behaviour between the growth behaviour of cold-worked (or cold-worked plus stress-relieved) material and annealed material are:

- a) the fluence index q in equation (2.1) is higher for cold-worked material, ranging from 0.28 to 1.0;
- b) there is no tendency towards saturation of growth at the higher fluences.

Typical growth-fluence plots for cold-worked material are given in Figure 4 and experimental values for the fluence index q are given in Table 2. Cold-work gives rise to a component of positive length change in the direction of working (rolling) (27) and the component of growth, q , increases with the amount of cold work (18,28).

2.2 Effects of Irradiation Variables

2.2.1 Effect of Neutron Flux

The effect of neutron flux intensity and flux distribution on growth has not been extensively studied experimentally.

* CANadian Deuterium Uranium
 + Stream Generating Heavy Water Ractor
 x Boiling Water Ractor
 † Pressure Water Ractor

However, the growth measurements of Murgatroyd and Rogerson (23) at 353K and 553K of polycrystalline zirconium, Zircaloy-2 and Zirconium-2.5wt%Niobium over the limited flux range of $5.3 \times 10^{17} \text{ n/m}^2 \cdot \text{s}$ to $11.6 \times 10^{17} \text{ n/m}^2 \cdot \text{s}$, (determined by ^{54}Fe monitors) suggests that beyond any initial transients, the total growth strain of a given material is determined only by the test temperature and neutron fluence and is independent of the time to reach that fluence. Irradiation growth appears to be directly related to the integral displacement damage and therefore will be caused only by neutrons above a certain threshold energy (about 40 eV in zirconium) (32).

2.2.2 Effect of Temperature

The early results showed very little effect of temperature on the growth of Zircaloy. Buckley (2) found the growth rate to decrease slightly when α -Zr was irradiated and measured at 77K as compared to results irradiated at 350K and measured at 290K. Harbottle (18) found the temperature dependence of irradiation growth in annealed Zircaloy-2 to be small in the temperature range of 77K to 553K. The instantaneous growth strain at 310K was slightly greater than that at 78K, the opposite to Buckley's findings. For annealed Zircaloy-2, Harbottle found that the growth rate approached zero as the fast neutron fluence approaches $\sim 5 \times 10^{24} \text{ n/m}^2$, for the temperature range of 77K to 553K which is consistent with most of the data presented in Figure 2, except for Adamson et al's results (70) at high temperatures.

Murgatroyd and Rogerson (23) measured the growth strains occurring in longitudinal 25% cold-worked Zircaloy-2 at 353K and 553K and noted an increase in the growth rate by a factor of around 2, as illustrated in Figure 5. The 353K data obtained an initial rapid transient in growth strain greater than that at 553K by about 2.9×10^{-4} , yet the higher temperature data has a higher growth rate and as a result overtakes the low temperature data at about $8 \times 10^{24} \text{ n/m}^2$.

Thermal cycling from 553K to 353K results (33) in a positive growth strain transient of 3.4×10^{-4} , Figure 6. The growth rate then equals the 353K growth rate which suggests that the prior growth rate at 553K had no effect. Upon raising the temperature back to 553K, a negative transient growth strain of 2.8×10^{-4} is obtained which removes most of the positive transient growth strain obtained earlier when the temperature was increased from 353K to 553K. The growth rate then resumes that of the 553K data. Three conclusions have been drawn from these observations: i) the transient growth strain is almost completely recoverable during temperature cycling under irradiation; ii) the characteristic linear growth rates at one temperature are unaffected by prior irradiation at the other; iii) growth strains achieved as a result of linear growth (rather than transient growth) at either temperature are not recovered during temperature cycling.

Adamson's (18) early results on recrystallized zirconium showed the fluence of irradiation temperature to be

small for fluences below $5 \times 10^{24} \text{ n/m}^2$ (E 1MeV) but significant at higher fluences $2 \times 10^{25} \text{ n/m}^2$. The maximum growth appeared to reach a peak at around 570K and then, it could be speculated that the growth would decrease with increased temperature. However, recently results by Adamson, Tucker and Fidleris (12) have found that the growth of both annealed and cold worked Zircaloy increased rapidly with temperature above 650K as illustrated in Figure 7. The increase in growth above 650K parallels changes in microstructure seen in irradiated Zircaloy by TEM (34). Above this temperature, the neutron damage defects, i.e., dislocation loops grow, interact and form dislocation tangles. The annealed microstructure becomes more like that of cold-worked material. Also, denuded zones have been seen at grain boundaries at 675K (35). As a result the growth in annealed Zircaloy changes from saturating to steady state behaviour similar to that of cold-worked material (10). This implies a change in growth mechanism away from that dominating at lower temperatures in annealed material. No systematic study of the temperature dependence of growth in cold-worked materials has been carried out at higher temperatures but Fidleris (12) speculates that the steady state rate should increase gradually with temperature up to about 600K and then more rapidly thereafter.

2.3 Effect of Material Variables

2.3.1 Microstructure

The rate of growth is largely determined by microstructure (12,13), i.e., dislocation density, grain size and

shape. Irradiation induced dislocation loops are thought to contribute to the growth strain during the early stages (13,36), but cannot fully account for the measured growth at high fluences (37). The pre-irradiation dislocation network is thought to contribute to growth by continuing growth at a steady state rate at high fluences (12,23,27, 36).

Much work has been done to characterize the irradiation induced defects, using TEM analysis. The damage created by neutron irradiation is not visible by TEM until the fluence reaches $\sim 2 \times 10^{23} \text{ n/m}^2$ ($E < 1 \text{ MeV}$) (37). At this fluence the damage, in the form of dislocation loops is greater than 2 nm in diameter (37). At low temperatures, ($T < 550 \text{ K}$) the dislocation loops appear as black spots which are electron-deficient rings whose strain fields result in diffraction contrast (5). At higher irradiation temperatures ($T > 550 \text{ K}$), larger unfaulted dislocation loops are seen, whose size and density increase with increasing fluence (37). The number of atom sites involved in the damage increases and then saturates as a fluence of $\sim 10^{25} \text{ n/m}^2$ ($E > 1 \text{ MeV}$), due to a slight decrease in the density of the visible defects while the size increases slowly. This is shown in Figure 8 from the results of Carpenter and Northwood (37) and Williams and Gilbert (38), confirmed by more recent results of Gilbert et al (34). The predominant dislocation loops lie in approximately equal number on the three possible $\{10\bar{1}0\}$ prism planes, having a Burger's vector of $a/3 \langle 11\bar{2}0 \rangle$ (5,7). The loops are not pure edge in character and can

tilt up to 30° from the $\{10\bar{1}0\}$ planes (39,40,41). A review of the data on Burger's vectors determination of dislocation loops is listed in Table 3, along with the condition and type of Zirconium-based metal, the source of irradiation, fluence levels, and vacancy/interstitial character (when included by the reference). There is some evidence of $\langle c \rangle$ component dislocation loops (41,42,43,45,46,58,59,63) created by various types of irradiation (i.e., neutron, ion and electron), their density being relatively very low (41, 42,63) and it is speculated that their presence is enhanced by impurities (42,63). In the "round-robin" experiment which was designed to characterize neutron irradiation damage in various Zirconium alloys, all participants agreed that no $\langle c \rangle$ component dislocation loops existed in any of the specimens examined. The corduroy structure (61), Figure 9 where dislocation loops aligned in rows having a displacement vector with a $\langle c \rangle$ -component illuded researchers (58,59,61,80) for some time until Kelly et al (51) proved conclusively that corduroy contrast is an artifact associated with local relaxation of the TEM thin foil specimen in the areas where there is a pronounced alignment of the $a/3\langle 11\bar{2}0 \rangle$ loops, and there is an excess of either vacancy or interstitial loops in each band. This was confirmed by Northwood et al (9) for zirconium, neutron-irradiated at 673K. As can be seen in Table 3 that the percentage of loops that are vacancy in character increases with increased irradiation temperature such that at irradiation temperatures of $\sim 673\text{K}$ there is an

excess of vacancy loops. At 773K there are a few large loops or none at all. Dislocation tangles still remain. By 873K there is no dislocation network left. It has not been possible to characterize the dislocation loops in zirconium alloys below ~550K (5), although post irradiation annealing has been used to coarsen the loops to a size at which the appropriate contrast experiments could be carried out (15,39). The results show that both interstitial and vacancy-type loops are present with an excess of the latter. This suggests that both types are present during irradiation at temperatures ~550K. It has been suggested that the vacancy loops are nucleated in the depleted core regions of the displacement cascades during neutron irradiation (68,71). Though recently, results of positron annihilation experiments (69) suggest that large concentrations of three-dimensional vacancy clusters are not created from the displacement cascades during neutron irradiation in zirconium. This suggests that the damage takes the form of discrete point defects and very small dislocation loops (36). At low temperature irradiation, the damage exists on such a fine scale that it is not readily characterized because of the oxide film on the zirconium TEM thin foil surface. Even in titanium, a h.c.p. material with similar irradiation properties as zirconium, but has less severe oxide film formation on the surface, the characterization of low temperature irradiation damage is difficult (68). The small loops are imaged at a given depth from the specimen surface in dark field under two-beam conditions at small positive deviations from the Bragg orientation (70). The 'butterfly'

type image is then compared to theoretical images created by a computer simulation technique and its vacancy/interstitial character determined (71). It is much easier to determine the character of larger dislocation loops (>2 nm) by using the inside/outside techniques (50,56,71,72,73), once the loop plane normal is determined. The safe orientation for loop characterization is in the $\langle 11\bar{2}3 \rangle$ which gives good contrast behaviour and reduces interstitial/vacancy loop mixup (56). During HVEM experiments the dominant $\langle a \rangle$ type dislocation loops, $\underline{b} = a/3\langle 11\bar{2}0 \rangle$ have been seen to nucleate on the $\langle 11\bar{2}0 \rangle$ planes in pure edge configuration and then reorientate during growth in order to reduce their energy by developing some screw character (68) and the lesser $\langle c \rangle$ type dislocation loops, $\underline{b} = a/6\langle 20\bar{2}3 \rangle$ (low dose) and $\underline{b} = a/3\langle 1\bar{2}13 \rangle$ (high dose), have been observed to appear during electron irradiation by glide from the basal plane (63). Also, at high irradiation temperatures (~ 773 K) the dislocation loop structures do not reach a steady-state size at high doses but coarsen to form dislocation networks (68,67) which could yield a change in growth mechanism as seen in Adamson's et al data (10) for growth of zannealed Zircaloy-2 at high temperatures 670K. Above this temperature region (>923 K) no irradiation damage exists during irradiation (67).

As we have seen, cold worked Zirconium-based alloys obtain a steady state growth rate at high fluences whereas the growth in annealed Zirconium tends to saturate at high

fluence ($\sim 10^{25}$ n/m², $E > 1$ MeV). Adamson (19) has shown that the growth rate increases as the degree of cold work increases. Holt (28) has analysed the effect of dislocation density, determined by x-ray line broadening measurements and electron microscopy (74), on the irradiation growth rate of cold worked Zircaloy-2 pressure tubes assuming the grain shape and texture dependence given by Holt and Ibrahim (27). The growth rate of pressure tubes varied with the dislocation density by the power of 0.82, i.e., $\rho_N^{0.82}$ (28). He then compared his data to results by Adamson (19) and found the dependence of growth rate on dislocation density to be similar to his own, substantiating this relationship.

Riley and Grundy (45) found for iodide Zirconium cold working 30% before neutron irradiation at 673K, reduced the mean size of the dislocation loops from 30-40nm for the annealed material to 20-30 nm for the cold-worked material. This observation is not unexpected since the straight dislocations in the cold worked structure could act as sinks for the irradiation-induced interstitials, thereby reducing the number available to plate out as dislocation loops. TEM evidence (75) of the damage structures in annealed and 20% cold-worked Zircaloy-2 after irradiation at 573K to a fluence of 1×10^{26} n/m² ($E > 1$ MeV) shows a similarity of damage structures in the two materials where there is no retention of any pre-existing dislocation structure in the cold-worked material. There was also no difference in the size or density of damage in the annealed and cold-worked

Zircaloy-2. Northwood (5) states that this is strong evidence for irradiation-enhanced recovery of pre-existing dislocations since merely holding the cold-worked material for an equivalent time out of flux at 573K would not recover the dislocation structure. Gilbert and Holt (11) state that the straight dislocations may be present but are obscured by the irradiation damage, since pre-existing $\langle c \rangle$ component network dislocations produced by cold working (108, 11) persist to high neutron fluences. The $\langle c \rangle$ component dislocations studied by Gilbert and Holt (11) were predominantly edge character before irradiation but changed to screw dislocations during neutron irradiation. This supports models (36,28) of irradiation growth requiring the presence of network dislocations to act as point defect sinks or paths for short circuit diffusion of point defects to grain boundaries in order to explain the high growth rate in cold-worked Zircaloy.

The contribution of grain boundaries is significant. Polycrystalline zirconium exhibits an order of magnitude greater strain than that of single crystals (14). A comparison of Carpenter et al (16) single crystal data, Figure 2 to the published growth in polycrystalline zirconium and its alloys, Figures 3,4, indicate that grain boundaries are important in the growth process. Almost all the proposed models of growth emphasize the importance of grain boundaries as sinks for point defects. The direction of strain when a point defect is absorbed at a grain boundary is the

direction of the grain boundary normal. Thus the growth rate increases as the grains become more elongated in a given direction, which occurs during the forming process when the grains are elongated in the direction of working and compressed in directions normal to the direction of working (76).

The effect of grain size on growth has not yet been well established (12). The only information (28) relating grain size to irradiation growth is that for pressure tubes operating in power reactors. Tubes with similar dislocation densities and a factor of two variation in grain size had similar growth rates. This evidence suggests that the effect is small.

2.3.2 Texture

Fabrication of anisotropic metals nearly always leads to the development of non-random textures, because of the preferred direction of deformation of individual grains during the forming process (76). Growth strain measurements depends linearly on texture, where the growth anisotropy factor, G_d , is related to the crystallographic texture by:

$$G_d = 1 - 3f_d^C \quad (2.2)$$

where f_d^C is the fraction of basal poles in the d direction. The values of f_d^C are derived from basal pole figures obtained by x-ray diffraction using the relationship (27):

$$F_d = \int_V \cos^2 \alpha_d \quad (2.3)$$

where V_α is the volume fraction of grains with their basal plane normals at an angle, α from the d direction. The experimental confirmation of this law can be found in the work by Ibrahim and Winegar (78). These authors, by assuming isotropic growth in the direction parallel to the basal plane and hence the above mentioned law were able to correlate data from Zirconium single crystals and from highly textured zircaloy rod with those from different Zirconium alloys with different metallurgical treatments. They observed a large scatter among experimental data and the main lack of correlation takes place for heavily cold-worked specimen. Murgatroyd and Rogerson (23,33) have found growth to be strongly dependent upon texture, whereas Madden (80) found no obvious correlation between texture and irradiation growth. Results, by Adamson (18) questioned the validity of the $(1-3f_d^C)$ law when recrystallized Zircaloy-2 tested at 339K with $f_c = 0.32$ gave a positive strain under irradiation while another specimen with $f_d^C = 0.338$ contracted under irradiation at 553K. Recently though, the results by Adamson et al (10) confirm the linear dependence of growth strain on the crystallographic texture as shown in Figure 10, for annealed and cold-worked-stress-relieved (3 hrs/740K) Zircaloy-2 slab material.

From the reasoning of Holt and Ibrahim (27), the growth anisotropy factor is given by:

$$G_d = 1 - f_d^C - 2A_d \quad (2.5)$$

where A_d is the grain boundary anisotropy factor for the "d" direction. Holt and Ibrahim have obtained reasonably good results using the growth anisotropy factor, G_d , given by equation (2.5) and this will be covered in more detail in the growth models, Section 2.6.

2.3.3 Alloy Composition

Work by Northwood (81,82) on a variety of zirconium alloys that were neutron irradiated at ~570K showed that increasing amounts of alloy addition in solid solution in the α -phase caused a reduction in the size and increase in the density of the damage (5), Table 4. Although the size and density of the damage varied with alloy content, the number of atom sites involved in the damage was essentially the same for a given fluence regardless of alloy content. This supports earlier evidence (83) of a Zr-2.5wt%Nb alloy in which the niobium in solution in the α -phase was varied from 2.7% to 1.1% by varying the heat treatment, where it was shown that after neutron irradiation at 573K the visible damage regions had a larger diameter in the α -phase which originally contained 1.1% Nb in solution than that originally containing 2.7%Nb. The excess solute was considered to restrict the growth of loops formed at the irradiation temperature.

Northwood (5) comparing TEM results of neutron irradiated zirconium alloys by Riley and Grundy (45) and Ells et al (35) at 673K with his own at 573K, concluded that Sn in solid solution was effective in restricting the

growth of dislocation loops at 573K irradiation temperature, but at about 673K, it lost its effectiveness whereas Niobium was seen to be equally effective at restricting dislocation loop size at both 573K and 673K, respectfully.

Recently, Buckley et al (65,66) have used HVEM at 1 MeV to study dislocation loop nucleation and growth in 99.99% Zirconium and a number of its alloys. It was seen that the saturated dislocation loop densities decreased with increasing irradiation temperature for all zirconium material at temperatures >573K. Increasing the niobium alloying addition from 0.1wt% to 2.5wt% (~0.1% to ~0.6% in the α -phase) increased the loop density (i.e., decrease the loop size) by almost an order of magnitude. Alloying additions of 0.15 wt%Fe and 0.15wt%Ni had no effect on the mean loop density at 700K.

Buckley's observations have led him to make three conclusions regarding the effect of alloying, which are:

- (i) the Sn present in Zircaloy acts as an atom-sized vacancy trap with a binding energy of 0.3eV; ii) the vacancy is more mobile ($E_V^m = 0.65\text{eV}$) than previously thought ($E_V^m = 1.2\text{eV}$), and iii) that the solute-interstitial binding energy is <0.1eV ($E_{bi} = 0.05$ for Sn in Zr-2 and $E_{bi} = 0.04$ to 0.09eV for Nb in 0.1wt%Nb and 2.5wt%Nb alloys respectfully). Thus irradiation induced point defects (i.e., vacancies and interstitials) are significantly trapped by oversized solutes (i.e., Sn and Nb) and are not much affected by the under-sized solutes (i.e., Ni and Fe).

The growth measurements by Murgatroyd and Rogerson (23) of polycrystalline zirconium, annealed Zircaloy-2 and annealed Zirconium-2.5wt%Nb illustrates the effect of alloy composition, Figure 11. The annealed Zircaloy-2 has the highest alloying additions in the α -phase (1.5wt%Sn) and shows a high initial growth transient followed by an early saturated growth. The annealed Zr-2.5wt%Nb having 0.6wt%Nb in the α -phase has a high initial growth transient but does not saturate at higher irradiation doses, continuing growth at a steady-state growth rate. The polycrystalline zirconium, being the most pure material has a longer duration in growth transient and eventually surpasses the growth strain of Zircaloy-2 and Zr-2.5wt%Nb. It is too early to see if the polycrystalline zirconium will saturate or continue at a steady state growth rate. It is expected to obtain a slightly higher steady-state growth rate than the Zr-2.5wt%Nb alloy. It is obvious that the alloying additions of Sn and Nb increase the initial growth transient by enhancing the nucleation and growth of dislocation loops. Sn increases the recombination of vacancies and interstitials by reducing the mobility of the vacancies. The annihilation of point defects in Zircaloy-2 has virtually reached a steady state giving an almost zero strain rate. The niobium alloying addition does not trap point defects as effectively as Sn atoms and allows net fluxes of interstitials and vacancies to be lost to various sinks, giving a steady-state growth rate. Polycrystalline zirconium, lacking alloying additions has a slower nucleation and growth rate of dislocation loops and has a continuous net loss of interstitials and vacancies

to various sinks resulting in a slow initial growth transient and a high growth rate.

Impurities have a similar effect on growth as alloying additions do. Zirconium and its alloys have a relatively high concentration of impurities. Fong and Northwood (84) have analysed impurities in Zircaloy-4 nuclear fuel sheathing material. They detected many second phase particles, cubic in nature, consisting of zirconium carbides, zirconium phosphides and free silicon. The effect of impurities on growth can be seen in single crystal material irradiated at 550K by Carpenter et al (15), Figure 2. The zone refined material, being the less pure, had a marked increase on the initial a-axis growth rate and a corresponding decrease in the duration of the transient, as compared to the purer, iodide refined Zirconium material. The effect of impurities are expected to enhance the nucleation of interstitial loops, increase recombination due to point defect trapping and, maybe, change the fraction of depleted zones that collapse to form dislocation loops.

Impurities have also been used to explain the stability of vacancy loops surrounded by interstitial loops and voids (42). Three items of circumstantial evidence favour the association of impurities with vacancy loops: i) Interstitial loops form 'rafts' while similarly sized vacancy loops do not. Since rafts form by glide, this implies that glide of vacancy loops is in some way impeded - possibly by impurity drag; ii) Faulted loops ($b = a/6\langle 20\bar{2}3 \rangle$) observed in less pure zirconium are invariably vacancy loops - another apparent

vacancy/impurity effect; iii) Large-vacancy ($b = a/3\langle 11\bar{2}0 \rangle$) loops in zirconium are often marked elliptical while similar sized interstitial loops are not. Since zirconium is virtually elastically isotropic, circular loops would be expected and hence the vacancy loops would be anomalous. The identification of the specific impurity responsible for the selective interaction with vacancy loops was not possible.

The only other observation of the effect of impurities on dislocations was by Bailey (85) who noted that in zirconium with high oxygen and nitrogen concentration, 0.1wt%, the straight dislocations appeared less jogged and more restricted to the $\{10\bar{1}0\}$ planes than zirconium material of higher purity.

2.4 Effect of Post-Irradiation Annealing

For annealed specimens Adamson showed that 100% of irradiation growth could be eliminated by suitable thermal anneals. Growth strains of $6-9.1 \times 10^{-4}$ measured during irradiation could be offset entirely by a length shrinkage during thermal annealing at 783K for 30 hours and that 73% to 87% of the growth strain could be recovered at an annealing temperature of 720K for 6 hours. Northwood and Causey (86) were able to show that for annealed Zircaloy-2 irradiated at ~ 573 K, annealing at 673K to 773K, the strain recovery was related to the annealing out of the damage. The effect of post-irradiation annealing was to increase the size of the damage and to reduce the density (Table 5). Post-irradiation annealing at 773K for 90 hours annealed

out all the damage. If we calculate the number of atom sites involved in the damage, N_d , we find that at 673K, although the size of the damage is increasing and its density decreasing, annealing times of more than 20 hours are required before N_d decreases. What appears to be happening up to 20 hours is that the point defects (vacancies and interstitials) are being redistributed into larger loops, with a consequence reduction in the overall density. After 20 hours at 400°C, annihilation of the defects occurs, reducing N_d and recovering the growth strain.

For cold-worked specimen, Adamson (18) showed that only a fraction of the growth strain could be recovered by post-irradiation annealing. As the % cold-work increased, the fraction of recoverable growth strain decreased. Results by Murgatroyd and Rogerson (23) support this observations. For both 25% cold-worked Zircaloy-2 specimen with measured growth strains of 7.3×10^{-4} and 11.72×10^{-4} , a recoverable strain of 2.5×10^{-4} at 553K was obtained. Rapid growth recovery occurred within the first few hours and then recovery decreased substantially with annealing time, Figure 12. Murgatroyd and Rogerson also report (23) that in reactor thermal cycling studies indicate that a strain in the range of 2.8 to 3.5×10^{-4} is recovered upon cycling between 353K and 553K.

2.5 Conservation of Volume

There is still some controversy about the growth strain being a constant volume process or whether it has

a volume component associated with it. Early evidence (87) suggested that volume changes were small, and made small contributions to growth. Intuitively, Parsons et al (3) assumed a volume increase for growth in their early model. Carpenter et al's (15) single crystal data of growth had a calculated volume increase of the same order in magnitude as the growth strain. Murgatroyd and Rogerson (13) measured a density decrease of $5.2 \times 10^{-4} \text{ kg/m}^3$ resulting in an isotropic volume strain increase of 1.7×10^{-4} and the small positive strain occurring at low doses has been taken as evidence of a volume increase (33).

For textured polycrystalline zirconium, Harbottle and Herbillon (88) reason that the absence of suitably orientated sinks for the point defects puts the constant volume process in question. Density measurements on cold worked and annealed Zircaloy-2 irradiated to $2 \times 10^{25} \text{ m}^{-2}$ ($E > 1 \text{ MeV}$) at 573K were determined using the standard immersion technique, indicating a significant volume increase ($\sim .1\%$) occurring during irradiation growth. They propose that the volumetric component is due to an excess of interstitials ejected from the uncollapsed displacement cascades and the increase in volume occurs during the early stages of growth due to the formation of depleted zones up to a steady state concentration. Beyond the lifetime of the depleted zones ($\sim 10^6 \text{ s}$ or 300 hours) (88), the swelling rate should cease. Using HVEM void density results (89), Harbottle calculates a volume strain increase of $\sim 10^{-3}$ at 573K with an average void size of $\sim 3 \text{ nm}$, too

small for TEM detection which is complicated by the presence of a high dislocation density.

The greatest problem in determining whether growth has a volume component, is the difficulty in measuring the growth strains in the three principal directions. Recently however, Adamson et al (10) have provided evidence for growth being a constant volume process up to 680K by taking growth measurements in all three principal directions, on annealed and 20% cold-worked-stress-relieved Zircaloy-2 materials. Also, results of positron annihilation experiments (69) suggest that large concentrations of three-dimensional vacancy clusters (Seeger zones) are not created from the displacement cascades in neutron-irradiated zirconium. The damage is therefore presumed to take the form of discrete point defects and very small dislocation loops (36). A volume component of growth may be associated with the dislocation cores of loops, formed during the early growth transient (23) and be completed once dislocation size and number (16), or more approximately, once the number of atom sites involved in the damage (5) has saturated.

2.6 Models

Buckley's model (2) was the first to account for irradiation growth in zirconium alloys, on the basis of his fission fragment irradiation experiments. He proposed that vacancy loops formed parallel to the basal planes and the interstitials precipitate as loops on the prism planes. Electron microscopy studies have since suggested

that this mechanism is not valid due to a lack of evidence for vacancy loops on the basal plane.

A modified Buckley mechanism was proposed by Kelly, (13) in which point defects migrate differentially to grain boundaries. In this model interstitials and vacancies migrate preferentially to grain boundaries aligned perpendicular to the crystallographic $\underline{a}$ and $\underline{c}$ axis, respectively. Stresses generated between grains as a result of the anisotropy of thermal expansion were suggested as the cause of defect partition. This mechanism is considered unlikely (37) since, once the stress from one thermal cycle has been relieved, further growth should drop to a low level, roughly that of a single crystal, which is insufficient to account for the measured growth values in polycrystals which are at least an order of magnitude larger than for single crystals (13).

Another mechanism, originally applied to creep (90) and then for growth was dislocation loop alignment in which there is a preferential alignment of interstitial dislocation loops, producing a positive strain in the direction of the Burgers vectors of the loops. Carpenter and Northwood (4) demonstrated that, at least for annealed Zircaloy-2 irradiated at about 570K, a mechanism of loop alignment was not viable for irradiation growth of polycrystals. In a growth strain calculation based on the observed dislocation loops and the assumptions that:

- 1) all loops have the same character (i.e., interstitial);

- 2) the specimen has 100% texture so that it behaves as a single crystal;
- 3) the loops all have the same Burger's vector.

The calculated strain was far less than the measured strain, particularly at large fluences. The calculation is an upper bound value and in practise, the true growth strain from dislocation loops would be much less, since there exists a mixture of vacancy and interstitial loops distributed evenly on all three possible $\{10\bar{1}0\}$ prism planes. The strain contribution from the vacancy and interstitial loops should cancel each other out. From the above considerations, it is considered unlikely that loop alignment is a viable mechanism for irradiation growth of polycrystalline zirconium and its alloys.

Carpenter and Northwood (37) suggested a very simple mechanism for growth based on the bias attraction between interstitials and dislocations. They suggest that the bias (which had previously been applied to void swelling behaviour in metals (91)) will cause an excess of interstitials to arrive at dislocations, leaving the residual vacancies to annihilate at neutral sinks such as grain boundaries. The dislocations climb by absorbing excess interstitials causing an expansion along the a-direction, i.e., in the direction of the Burger's vectors. The annihilation of residual vacancies at each grain boundary causes a contraction perpendicular to the boundary. The result is a net expansion along directions in the basal plane and a contraction parallel

to the c-axis. Qualitatively, this mechanism accounts for the faster growth of polycrystals, as compared to single crystals. In addition, increasing the dislocation density would be expected to increase the growth rate, which agrees with the experimental observations (18,28). There is good TEM evidence of grain boundaries acting as a sink for point defects resulting from irradiation. In Ni irradiated by 4 MeV Ni ions (92) grain boundaries are shifted during ion irradiations and voids which formed during irradiation are absent near the grain boundaries, forming a denuded zone. In zirconium, TEM evidence (35) suggests a net flux of vacancies to the grain boundaries. In a region next to the grain boundary, the dislocation loops are absent when the loop character is primarily vacancy. This observation is more prevalent at higher irradiation temperatures ($\sim 400^{\circ}\text{C}$) where the majority of loops are vacancy in character.

Dollins (93) proposed a multimechanistic model where growth comes from preferred nucleation and growth of interstitial loops, climb of dislocations with preferred Burger's vectors, depleted zone formation and point defect absorption at subgrain and grain boundaries. The vacancy sinks in his mathematical model of growth are straight dislocations, dislocation loops, depleted zones and grain boundaries. For annealed materials, Dollins considers the loops to be the main source of growth with lesser contributions coming from grain boundaries and straight dislocations. Using his model, the growth values have been found

to be in excellent agreement with measured experimental data. However, there are a number of flaws in the Dollins model, the main ones being that his choice of loop size at high fluences is much larger than experimentally observed (5,56,86), and his complete neglect of the contribution of the vacancy loops which are present, particularly at the higher irradiation temperatures. The growth was predicted to come from mainly interstitial loops which grew continuously despite evidence to the contrary (94). As a result, the calculated growth rate does not decrease with dose as observed in practice for annealed material.

For cold worked material, Dollins considers the climb of straight dislocations to be the dominant mechanism at all temperatures, with minor contributions coming from grain boundaries and dislocation loops. The predicted values for interstitial loop radius do agree well with experimentally measured values. As well, the growth predicted is in good agreement with some measured growth values (95). A model such as Dollins (93) would seem to be the logical answer since any one of the sources for point defect annihilation is unable on its own to account for the measured growth.

Adamson (18) looked at the mechanisms for growth and agreed with Dollins (93) that multi-mechanisms are operating. He assigned the major contributions to growth for annealed materials as depleted zones and vacancy loops (rather than interstitial loops). The vacancy loops seem to fit the experimental observations better, both from a standpoint of TEM observations and growth measurements.

His assessment of the irradiation growth mechanisms for annealed Zircaloy-2 is reproduced in Table 6, with the exception of corduroy, a thin foil artefact (51), and, including comments as to their relative contributions.

A reasonable mechanistic growth model has been proposed by Murgatroyd and Rogerson (23) to explain the behaviour of annealed and cold-worked Zircaloy-2 at 353K and 553K (78,96,97,98). This model proposes that in the early stages of irradiation sub-microscopic interstitial clusters form on prism planes giving anisotropic, texture dependent growth. As the concentration of clusters, vacancies and depleted zones increases with irradiation substantial recombination occurs and growth by this mechanism effectively ceases. In annealed Zircaloy-2, it is proposed that this leads to virtual saturation of growth, but in cold worked material it is postulated that growth can continue by preferential absorption of interstitials and the drift of vacancies to an alternative sink. In addition, it is probable that a small volume increase results from the formation of a high vacancy concentration and the volume change associated with the core of dislocation loops. For the annealed Zircaloy material, the saturation of growth is the result of vacancy trapping by Sn atoms, which enhances the recombination of interstitials and vacancies.

Although the mechanistic model (23) has been proposed, no detailed calculations have as yet been performed to verify the model.

There have been many models of growth based upon reaction rate theory and the bias of point defects to sinks. The general form of the rate theory equation for a point defect is given by:

$$dc_x/dt = P_x - R - \sum_K J_K^x \quad (2.4)$$

where dc_x/dt is the change in defect concentration with time, P_x is the production rate of x type defect, R is the rate of recombination and $\sum_K J_K^x$ is the loss of the x type of point defect to a K type of sink.

Fainstein-Pedraza, Savino and Pedraza (FPSP) (99) proposed a simple model for calculating the irradiation growth of polycrystalline zirconium-base alloys. The model has two point defect sinks, namely the lattice dislocation network and grain boundaries. The dislocation content, having a $\langle 11\bar{2}0 \rangle \{10\bar{1}0\}$ character is considered constant throughout irradiation. The model allows the dislocation loop radius to increase with irradiation dose. The steady state strain rate is assumed to contain a large contribution from the continued growth of interstitial loops. Both are in contradiction with experimental studies where both interstitial and vacancy loops are present and their size saturates with increased neutron fluence. The specimen texture is included by considering it as the result of a limited number of grain orientations. The mutual recombination of vacancies and interstitials created from the primary knock-on collision during irradiation is kept constant and it is assumed that mutual recombination occurs spontaneously

only when an interstitial and vacancy become nearest neighbours. This recombination distance is demonstrably low and it is thought (36,28,100,101) that a more reasonable recombination distance of between 30 to 50 lattice sites is adequate. In the rate equations bias factors are used for determining the probabilities for capture of each defect type by fixed sinks. The agreement between theoretical calculations and experimental values of cold worked specimen material are reasonably good when using an interstitial-vacancy bias factor ratio of 1.2, i.e., dislocations have a 20% higher attraction for interstitials than vacancies. Comparison of theoretical calculations with experimental data for annealed specimen material is poor since the dislocation loop size increases continuously with dose and thus no saturation of growth is attained. In a modified model presented by Pedraza and Fainstein-Pedraza (102,103), a better fit is attained for annealed material by varying the interstitial loop population and by introducing another type of dislocation loop in their model. They use dislocation loops having both a prismatic habit plane $(10\bar{1}0)$ and $\frac{1}{3}\langle 11\bar{2}0 \rangle$ Burger's vector and having a basal habit plane (0001) and $\frac{1}{6}\langle 20\bar{2}3 \rangle$ Burger's vector. The probability that either intrinsic loop type be produced is assumed to be equal. This behaviour again is not supported by experimental evidence (5). The bias factor for loops varies as a function of loop radius. In a recent publication for cold-worked zirconium-based alloys, Fainstein and Fainstein-Pedraza, have again included small

vacancy loops with a basal habit and a $\frac{1}{6}\langle 20\bar{2}3 \rangle$ Burger's vector. The theory is modified from cold worked material to fit annealed material (103) by assuming grain boundaries contribute very little to growth and there is a low dislocation content controlled by a high self-diffusivity coefficient leading to a saturation in growth. The model has a better correlation between calculated values and experimental observation than previously reported (99). Pedraya and Fainstein-Pedraza (102) then developed their rate theory approach of irradiation induced dimensional changes over a range of irradiation temperatures. The calculated values fit the high ($>535\text{K}$) and intermediate ($\sim 535\text{K}$) growth data of Adamson (18) and Murgatroyd (97) but, the rate theory could not fit the rapid saturation of growth ($\sim 2 \times 10^{24} \text{ n/m}^2$) at low temperatures ($<350\text{K}$). Pedraza and Fainstein-Pedraza (102) question whether any rate theory approach based upon the annihilation of point defects by means of diffusional processes can yield the experimentally measured strain versus dose behaviour at low neutron irradiation temperature.

Savino and Laciana (104) modified the FPSP model to include irradiation enhanced creep with the irradiation induced growth component of zirconium alloys. The specimen texture is included by considering it as the result of a limited number of grain orientations which allows them to solve a system with a finite number of equations for the total strain of the specimen.

They include the internal stress created by irradiation induced dimensional changes of the material which, they feel, may influence the grain growth rate and hence the total specimen distortion rate. Two new aspects included with respect to the FPSP theory are trapping of point defects and stress effects via the Stress Induced Preferential Absorption (SIPA) of point defects.

Brailsford et al (91,105) and Bullough and Wood (106) have set up a system to approximate irradiation induced dimensional change in metals and they have applied the system to zirconium and its alloys (106). The system is a set of discrete sinks, surrounded by a 'lossy medium' which is an infinite region in which defects are generated and lost to many types of sinks at rates which vary spatially and are proportional to the local defect concentration. The model employed is one in which the concentration of each type of point defect is assumed constant throughout the body and their losses to the various sinks as well as their annihilation by recombination are assumed to occur homogeneously. The rates at which these various losses occur involve the solution of a boundary-value diffusion problem where an individual sink of a particular type is represented by its actual geometry and size. The sinks used are interstitial loops, the dislocation network and vacancy traps where their bias factor varies as a function of relative sink strength. Dislocations have been given an interstitial-vacancy bias factor ratio of 1.25, slightly higher than FPSP's (99) estimate.

Wiedersich (107) applied a similar model for the bias attraction of point defects to sinks, using a Wigner-Seitz cell approach and surrounded the sink with a sink-free region, within which point defects are generated, whose outer boundary was chosen to give the same cell volume as the average volume per sink in the actual medium and no flux was allowed to cross the outer boundary.

Both of these approaches are approximations to the "real situation" in which many sinks of various types and geometries are spatially distributed, each with its own diffusional fields of both vacancies and interstitials which has generally been considered too complex to analyse directly (29). The bias factors are usually assumed to be kept constant and several attempts have been made to estimate their magnitude theoretically but all are seen to involve errors or physically unrealistic assumptions (100). The whole question of bias factors has come under scrutiny, and Nichols has concluded that any bias factors obtained up to this point must be considered empirical and there is a real need to obtain quantitatively reliable estimates of these all important bias factors (90,100). MacEwan and Carpenter (36), offer a very good review of irradiation growth behaviour, radiation damage structures, growth parameters necessary for the calculations, and a comparison of various modelling procedures used by others. They have attempted to develop a realistic model that relates to both measured growth rates and microstructural data. The calculations have been based on what they consider to be reasonable and justifiable estimates of thermodynamical

properties of the point defects in zirconium. Table 7, lists the values of the parameters used in the calculations. Sinks for point defects are grain boundaries, aligned and not aligned edge and screw dislocations, and aligned interstitial and vacancy loops. The sinks are not fixed during neutron irradiation and as a result there is irradiation enhanced recovery of intrinsic dislocations which is supported by experimental evidence (53). There are three defect types, namely vacancies, interstitials and free solutes which make three defect complexes, namely divancies, solute-vacancy and solute-interstitials. Recovery of the dislocation network is assumed to be controlled by climb of edge dislocations. Because of the imprecise knowledge of the bias factors, MacEwan and Carpenter consider that they are better treated as disposable parameters. In their calculations, the network dislocations are characterized by their density, edge to screw ratio and by the anisotropy of the Burger's vector distribution. The bias factor for dislocation loops is given as a function of loop radius, written as:

$$z_L^i = z_D^V + f(r_L)(z_D^i - z_D^V)$$

where $f(r_L)$ is given as a sigmoidal, decaying exponential function with increasing loop radius such that for small loop radii ($<3\text{nm}$), the bias is high for interstitials annihilating at loops. As the loop size increases the bias factor for interstitials to loops quickly decreases to become equal to the vacancy bias factor, thus limiting loop

growth. In the MacEwan and Carpenter model (36), a new postulate is used to describe the behaviour of network screw dislocations. It is proposed that they receive a net flux of vacancies which can be either annihilated at the screw dislocations, acting as a perfect sink, forming dislocation helices (model 1) or, the excess vacancies do not annihilate at the screw dislocations but migrate by pipe diffusion to the grain boundaries, thus short circuiting the vacancy path towards grain boundaries (Model 2). More realistically, only a fraction of the net vacancy flux arriving at screws would pipe-diffuse to the grain boundaries since some would be annihilated, either directly on the screws or indirectly by pipe diffusion to edge segments which would be receiving a net flux of interstitials. Also, the net flux of vacancies being piped to grain boundaries would be expected to decrease with increasing dislocation density. Model 2 has recently been supported by experimental evidence (43) where dislocations in cold worked Zircaloy-2 were seen to change from edge character before irradiation to screw character after irradiation in which they have not formed helices and hence have not absorbed defects. For these two models, a comparison is made between the calculated growth rates and experimental data (97) for cold worked Zircaloy-2 at 550K, Figure 14. Dislocation loops play no part in the growth which would be expected to cause an underestimate of the initial growth strain but have little effect on the long term behaviour. Initially, the network dislocation density is $10^{14}/\text{m}^2$, containing 75% of the network edge and 25% screw

with 75% of the Burger's vectors aligned in the measuring direction. For screw dislocations the bias for vacancies and interstitials is considered equal and a 3.5% preference is given for interstitials arriving at the edge dislocation network. In Figure 13, curve 1 was obtained assuming that screw dislocations act as perfect sinks forming dislocation helices (Model 1) and allowing irradiation-enhanced recovery of the network. Only two types of point defects are used, namely interstitials and vacancies. Curve 2 illustrates the effect of screw dislocations acting as pipes (Model 2) rather than sinks. Curves 3 and 4 show the effect of adding divancies and solute trapping on the calculated growth rate. The long term change in growth rate is a result of the network density changing with dose. Even though the growth rate is higher when using Model 2 than when using Model 1, it is still about a factor of 3 lower than the rate measured experimentally. Curve 5 illustrates the effect of having the dislocation density remain constant with no irradiation enhanced recovery of the network which is the common practise for most models (93,99,106), for which there is no long term transient to a decreased growth rate. From the comparison of the experimental data measured by Murgatroyd and Rogerson, MacEwan and Carpenter conclude that their model is incomplete and suggest four possible modifications in an effort to create a more realistic model. They are:

- 1) irradiation-enhanced recovery of the network;
- 2) short-circuit diffusion of vacancies along

- screws to grain boundaries;
- 3) annealing of mobile divancies;
- 4) solute trapping.

Also, in recent experimental work, Woo et al (108) and Gilbert and Holt (11,43) have shown that zirconium and its alloys may contain significant densities of dislocations having c-component Burger's vectors. Since the annihilation of point defects at these dislocations provide an additional component of strain along the c-axis it is important that they be included in future growth calculations. Adams, Clevinger and Hirth (109) have suggested an upper-bound evaluation technique for calculating the interactive creep and growth in textured zircaloy. The model uses a "crystallite orientation distribution function", (CODF) which is a stress-strain weighting function of crystallites found in a particular direction. Adams et al's model accounts for grain shape and size effects, as well as non-uniform dislocation densities arising from variable fabrication history. The internal strains are shown to interact with those dependent on applied stress in a manner which alters the constitutive behaviour yielding a growth affected local stress state. This stress state is distinguished from the stress component creating creep. The Holt-Ibrahim model (27) is an empirical model used to calculate the combined growth and creep rates of zirconium-based pressure tubes. Their model attributes growth due to the generation of equal numbers of vacancies and interstitials by fast neutrons,

by the condensation of point defects to sinks having anisotropic distributions because the microstructures are anisotropic. The partitioning of point defects occurs in the absence of applied stress due to the interaction between large dilation strain fields of interstitials and the long range strain fields of straight dislocations and dislocation loops. Since vacancies have a much weaker strain field, excess vacancies precipitate at sinks without long range strain fields such as clusters, voids, grain boundaries and free surfaces. The combined growth rate, $\dot{\epsilon}_g$ and creep rate $\dot{\epsilon}_c$ are given by (110):

$$\dot{\epsilon}_T = \dot{\epsilon}_c + \dot{\epsilon}_g \quad (2.6)$$

$$\dot{\epsilon}_g = K_g \phi G_d \quad (2.7.a)$$

$$\dot{\epsilon}_c = K_c \phi \bar{\sigma} C_d$$

where K_g and K_c are the measured specific growth and creep rate constants which depend on metallurgical structure and temperature, respectively, G_d and C_d are the growth and creep anisotropy factors in the "d" direction, respectively, ϕ is the flux and $\bar{\sigma}$ is the normalized applied stress.

The creep anisotropy factor, C_d depends on texture. The growth anisotropy factor G_d , is derived on the assumption that growth results from climb of dislocations with $a/3\langle 11\bar{2}0 \rangle$ Burger's vectors absorbing an excess of interstitials and causing expansion in the $\langle 11\bar{2}0 \rangle$ directions,

and a net deposition of vacancies at grain boundaries which causes contraction in the direction of the grain boundary normal. As a result of Holt and Ibrahim's (27) reasoning,

$$G_d = 1 - f_d^C - 2Ad$$

where f_d^C is the resolved fraction of basal plane normals in the "d" direction and Ad is the grain boundary anisotropy factor in the "d" direction. For grains with an approximate rectangular shape and with mean intercept grain sizes in the three fabrication directs of d_R , d_T , and d_L , and in the "d" direction of d_d , Ad is given by (114):

$$Ad = \frac{1}{d_d} / \left(\frac{1}{d_R} + \frac{1}{d_T} + \frac{1}{d_L} \right)$$

where Ad equals $1/3$ for equiaxed grains.

From these relationships, G_d and the growth rate increases as the grains become more elongated in the grain direction, "d". The Holt-Ibrahim model has proven to have the ability to accurately predict the growth and strain rates of pressure tubes (28). This model should be used as an empirical tool for predicting pressure tube behaviour until a well understood, mechanistic model can be applied.

Some researchers (97,12) prefer to mathematically model the growth data by using the equation:

$$G = K_1 + K_2 \phi t$$

where the growth G , as a function of neutron irradiation

dose is a straight line with a slope K_2 and intercept, K_1 . The intercept value can be used to account for the effect of residual stresses during the early growth period (12). The correlation of experimental data (97, 78) using this equation is usually better than using the exponential equation given earlier, as:

$$\epsilon = K(\phi t)^q$$

2.7 Experimental Methods to Measure Irradiation Growth and Creep with Particular Emphasis on the Cantilever Beam Method

The existence of irradiation growth in zirconium when first established by Buckley (2) was measured by the linear expansion and change in curvature of thin foils of annealed zirconium irradiated by fission fragments, Figure 14. Later, Harbottle (17) measured the differential growth strains of irradiated longitudinal and transverse specimens by welding the specimens together at the ends to make a 'bimetal' strip. The differential growth rates were measured by the change in curvature of the specimen during neutron irradiation.

In 1978, Parsons and Gilbert (113), irradiated a Zirconium thin foil by 100 KeV oxygen ions, in situ, (114, 115) in a Philips 300 electron microscope, permitting the simultaneous irradiation and observation of the microstructure. They observed (113) the formation of irradiation damage, growth of surface oxide and bending of the thin foil around the edges of the perforated hole, Figure 15, due to the 100 KeV oxygen ion irradiation. Recently, Parsons, Hoelke and

Gilbert developed an experimental method to measure the irradiation-induced deflection of thin foil (~0.127 mm thick) zirconium cantilever-beam type specimens bombarded by 100 KeV neon ions, Figure 16. These specimens show a 'bimetal-type' behaviour when irradiated.

The 100 KeV neon ions penetrate to a depth, d , resulting in a deflection, δ of the free end of the cantilever-beam specimen. 100 KeV neon ions are used because they produce a primary recoil spectrum close to that produced by fast neutrons (3,121). The deflection, δ has been partly attributed to irradiation induced growth of the damaged layer, d as the result of the interaction between the neon ion projectiles and the zirconium target material. A short review of the neon ion-zirconium target interactions is given in Appendix A.

The 100 KeV neon ion damage profile in zirconium has been calculated to have a profile as shown in Figure 17, where the ion damage layer, d is considered to be equal to twice the straggling of the damage distribution (~ 100 nm), centred at 81 nm below the ion irradiated surface (3, 121).

From simple beam theory (4) the deflection, δ of the free end of the cantilever beam of length, ℓ and thickness t resulting from a strain, ϵ and a stress, σ on the damaged layer of thickness, $d \ll t$ is given by:

$$\epsilon = \frac{t\delta}{\ell^2} \quad (2.10)$$

and,

$$\sigma = \left(\frac{1}{1-\nu}\right) E \epsilon \quad (2.11)$$

where E is Young's Modulus and ν is Poisson's Ratio.

Since a purely theoretical model for growth of zirconium and its alloys has not yet been able to account for the measured growth, the Holt-Ibrahim model (27) which includes both irradiation-induced growth and creep has been applied.

The Holt-Ibrahim model (27) is suitable for analyzing the deflection of the cantilever-beam specimens as there are three mechanisms assumed responsible, namely: 1) a volume expansion $\Delta v/v$ of the neon ion damaged layer due to the physical presence of implanted ions; 2) a structurally dependent expansion or contraction of the ion damaged layer due to irradiation growth; and 3) a stress which induces creep, opposing the dimensional changes in the ion damaged layer, generated by the undamaged bulk of the cantilever beam. Saturation of the deflection occurs when this opposing stress is equal to the net stress arising from neon ions and irradiation growth. The total strain rate, ϵ_t of the damaged layer becomes,

$$\epsilon_t = \epsilon_{\text{Neon Ions}} + \epsilon_{\text{Growth}} - \epsilon_{\text{Creep}} \quad (2.12)$$

In reference (3), Parsons developed a relationship between the deflection of the cantilever-beam and the fractional volume change, $\Delta v/v$ due to the neon ion irradiation damage. The same reasoning can be used for the expansion of

the zirconium lattice due to the presence of neon ions, (see Appendix B), from which the strain rate due to the presence of neon ions is given as (125):

$$\dot{\epsilon}_{\text{Ne}} = 1/3 \frac{A}{t} \dot{\phi} \quad (2.13)$$

where A is the volume expansion per neon ion, t is the thickness of the specimen and $\dot{\phi}$ is the neon ion flux.

The Holt-Ibrahim model reasons that the growth strain rate is given by:

$$\dot{\epsilon}_g = K_g G_d \dot{\phi} \quad (2.7)$$

and the creep strain rate by:

$$\dot{\epsilon}_c = K_c C_d \sigma \dot{\phi} \quad (2.8)$$

where the growth and creep rate constants, K_g and K_c depend on metallurgical structure and temperature, the growth anisotropy factor, G_d depends on texture and grain shape, and the creep anisotropy factor, C_d depends on texture.

These quantities have been defined earlier in Section 2.5.

The proposed growth and creep anisotropy factors for the cantilever-beam specimen are determined in Appendix C.

The growth anisotropy factor G_d is given as:

$$G_d = \frac{P_d}{2} (K+1) - K \cdot A'_{dg} - A_{dF} \quad (2.14)$$

where, P_d is the prism pole texture coefficient in the 'd' direction, A'_{dg} is the grain boundary anisotropy factor, A_{dF} is the free surface anisotropy factor and K is a geometrical constant.

The creep anisotropy factor is given as:

$$C_d = \frac{\sigma_{\perp}}{\sigma_{\parallel}} (\epsilon_{\perp\perp} + \epsilon_{\perp\parallel}) + \epsilon_{\parallel\perp} + \epsilon_{\parallel\parallel} \quad (2.15)$$

where the stresses σ_d are the normalized and component stresses in the 'd' direction, $\parallel$ means parallel to the cantilever-beam length, $\perp$ is in the cantilever-beam width direction and ϵ_{dd} are the texture dependent strains in the 'dd' direction. A biaxial stress condition is operative for creep during the irradiation of the cantilever-beam specimen.

From equations 2.10, 2.11, 2.12, 2.13, 2.7 and 2.8, the rate of change of the deflection of the free end of the cantilever-beam as a function of neon ion flux, metallurgical parameters and irradiation conditions is given by:

$$\frac{t\dot{\delta}}{\ell^2} = \frac{1}{3} \frac{A}{t} \phi + K_g G_d \phi - K_c C_d \frac{E}{(1-\nu)} \frac{t}{\ell^2} \delta \phi \quad (2.16)$$

Upon integrating this equation (Appendix D), the deflection of the cantilever-beam can be written as an exponentially decaying function,

$$\delta = C_1 - C_2 \exp(-C_3 \phi) \quad (2.17)$$

where,

$$C_1 = \frac{\frac{A}{3t} + K_g G_d}{K_c C_d \frac{E}{(1-\nu)} \frac{t}{\ell^2}}$$

$$C_2 = C_1 - \delta_0$$

$$C_3 = K_c C_d \frac{E}{(1-\nu)} \frac{t}{\ell^2}$$

where it is shown that the saturation in the deflection of the cantilever-beam specimen during neon ion irradiation is due to the texture dependent creep component. Also, a non-zero deflection, δ_0 at $\phi = 0$ has been introduced to account for any early transient growth due to stress relieving. The correlation between the 100 KeV neon irradiation and the fission neutron irradiation of zirconium has been determined by Parsons (125) (Appendix E). The resulting correlation factor between the two types of irradiation is given by:

$$\phi_{\text{neutron}} = 1.44 \times 10^4 \phi_{\text{neon ion}} \quad (2.18)$$

This correlation factor or one close to this value should be used in the analysis of the cantilever-beam results when comparing the experimental 100 KeV neon ion growth and creep rates to the neutron irradiation data collected by others.

CHAPTER 3

Experimental Details

3.1 Ion Irradiations

The 100 KeV neon ion irradiations were performed in a mass separator similar to the one shown in Figure 18, with a swept beam of doubly charged neon ions (Ne^{++}) at 50KeV with measured currents of up to $3\mu\text{A}/\text{cm}^2$ (9.4×10^{16} ions/ $\text{m}^2\cdot\text{sec}$) at a pressure of 1.5×10^{-5} Pa in a 'cyromet' trapped target chamber. The neon ion beam had a one minute sweep over four and sometimes two specimens mounted parallel to each other as shown in Figure 19.

The cantilever-beam specimen were irradiated at room temperature but due to ion beam heating the specimen temperature increased to 325K while the swept beam was on the specimen and cooled to 303K while the beam was off the specimen, (these temperatures were measured by 0.076 mm chromel-alumel thermocouple wires spot welded to the back surface of a dummy specimen). The specimen temperature has been taken as the average temperature of 314K.

The deflection of the specimens were measured outside the irradiating facility at room temperature (293K) after an incremental ion dose. The displacement of the specimen's free end resulting from the dimensional changes in the ion damaged layer was measured by mounting the specimens on an optical bench and focusing a He-Ne laser light onto mirrors attached to the specimen holder and the free end of the

specimen. The mirrors had a diffraction grating on their surface which gave a diffraction pattern on a wall 3.5 meters away, Figure 20. The mirror attached to the specimen holder remained fixed and was used for reference positioning. The displacement of the free end of each specimen was then determined by the change in angle required to re-position the mirrors so that their diffraction pattern (position 2 in Figure 20) coincided with the places they occupied before irradiation, (at position 1 in Figure 20). The specimens in their holder could be rotated by two micrometer screws magnetically coupled to the optical bench mounting apparatus giving 2-axes tilt and reproducibility of this re-positioning of $\pm 2.5 \times 10^{-6}$ m.

A comparison of the cantilever-beam specimen's microstructure before and after irradiation was made possible by electron microscope examination in a JEOL 100CX⁺ and Philips EM300G* microscopes using a double and single axis tilt stage respectively, both at 100KeV. The resolution limit for observing the irradiation damage in both microscopes was about 2nm.

3.2 Specimen Preparation

3.2.1 Cantilever-Beam Specimens Including Mirrors

The polycrystalline zirconium specimens were taken from an annealed rolled sheet as shown in Figure 21, having ~0.127

⁺ University of Windsor, Windsor, Ontario.

^{*} Atomic Energy of Canada Ltd., Chalk River Nuclear Laboratories, Chalk River, Ontario.

mm thickness. They were spark machine cut into cantilever-beam specimens 40 mm long by 2.5 mm wide.

All other specimens, Zircaloy-2, Zr-2.5wt%Nb and XL Alloy came from pressure tubing stock, Table 8. The longitudinal and transverse specimen were cut using a slow speed diamond saw from sections of pressure tubes parallel and perpendicular to the tube length, Figure 22, providing specimen blanks 45 mm long by 0.3 mm thick. A grinding jig was used to mechanically thin the specimen on silicon carbide papers to a thickness of ~0.15 mm, ensuring parallel specimen surfaces by placing 3 lead bricks and a rubber stopper on top of the specimen when being set in wax on the grinding jig surface. The specimens were then spark cut into cantilever-beam specimens, having dimensions of 40 mm long by 2.5 mm wide. A 1.2 mm diameter hole was spark cut at one end of all cantilever-beam specimens enabling them to be held by a brass plate and screwed on the cantilever-beam specimen holder, Figure 19.

The cantilever-beam specimens were then chemically polished to a thickness of ~0.125 mm in a acid bath containing 3.5% H F., 39% HNO_3 and 57.5% H_2O at room temperature for polycrystalline Zirconium and Zircaloy-2 specimens and 10% HF, 30% H_2SO_4 , 30% HNO_3 ; and 30% H_2O at 53°C for the Zr-2.5 wt%Nb and the XL Alloy. The specimens were immediately washed in water, then alcohol and finally air blown dry to produce a shiny surface.

The low-temperature-irradiation mirrors were made from

glass slides, cut into 1.25mm^2 squares. TEM microscope grids were placed on their top surface and aluminum evaporated onto them, leaving a diffraction grating Figure 23 which produces a diffraction pattern when reflecting laser light.

The high-temperature-irradiation mirrors were made from a 3 mm diameter, high-carbon steel rod, sliced into thin sections and then polished to a 0.05 alumina grit size. Carbon was evaporated onto their surface enabling them to reflect laser light in the same manner as the low-temperature-irradiation specimen, Figure 24. The low-temperature-mirrors were glued on with epoxy and the high-temperature-mirrors were spot-welded on, from the back of the cantilever-beam specimen to the back of the mirror.

The surface of the mirrors had a smoothness close to that of the wavelength of light, which ensures the reflection of laser light as a diffraction pattern.

3.2.2. Specimens for TEM Investigation

After the cantilever-beam specimens were irradiated, TEM specimens were prepared to study the nature of the irradiation damage. To study the irradiation damage normal to the irradiated cantilever-beam surface by TEM, the initial thinning process required placing the irradiation damage surface ($30 \times 2.5 \text{ mm}$) protected by wax onto a glass slide. The back surface was acid etched using the same etchants as before until there was perforation somewhere through the cantilever-

beam specimen. The specimens were immediately rinsed in water, dried and small ~3 mm diameter areas were covered with wax, protecting this material from further acid etching which left these areas as small discs suitable for the further TEM specimen preparation stages. The final thinning process involved electro-chemically thinning the specimen in a 5:1 ethanol-perchloric acid solution at ~240K, having an electrical potential of 18 volts. The process was stopped once perforation of the disc occurred, after which the specimens were rinsed in alcohol and then dried in air on filter paper. The TEM specimen thinning apparatus which was similar to that developed by others (133,134) is shown in Figure 25 and a typical TEM thin foil specimen is shown in Figure 26. Many different methods were tried to produce TEM thin foil specimens perpendicular to the irradiation damaged surface (perpendicular to the Neon ion beam direction). Such specimens were required so as to determine the irradiation damage depth (~100 nm) and distribution. Stereomicroscopy (135, 65) of the depth distribution in the normal direction samples could not be used because of the small size and high density of the 100KeV Neon ion damage. No standard technique could be followed since such measurements had never before been made in hcp zirconium-based materials. In techniques used for (136) nickel, copper (137) and steel specimens, the irradiated surface has been protected by electro-chemically depositing a f.c.c. metal into it and then preparing TEM thin foil specimens in the conventional manner, as discussed previously, waiting until perforation occurs at

the base-metal/deposited metal interface. For semi-conductor materials (138,139,140,141,142,143), e.g., silicon and germanium, the irradiated surface has been protected by using a silver-based epoxy*. The specimens have been initially mechanically thinned to a thickness of $\sim 50\mu\text{m}$ and then, the final thinning was accomplished by ion-beam sputtering of the surface until a hole is perforated or close to perforated at the specimen/silver epoxy boundary, producing a TEM thin foil specimen of the irradiation damaged layer.

Since a technique had not been developed for depositing metal onto a zirconium alloy surface (apart from very recently when nickel was electrolessly deposited (141)), the major experimental thrust during this master's thesis work concentrated on the silver epoxy technique to protect the irradiation damaged layer. The silver epoxy, epotek H20S, was a smooth flowing paste having a non-critical hardener to resin mix ratio of 1:1, also having good conductivity ($1-3 \times 10^4 \text{ ohm}^{-1} \text{ cm}^{-1}$, volume conductivity), setting at 90°C after a few hours and not gasing off at temperatures $\leq 400^\circ\text{C}$. Once set the epoxy's mechanical properties were like that of ceramic material, i.e., brittle, having a reported lap shear strength of 10 MPa. The chemical properties of the silver epoxy were suitable for making thin foil specimens. The epoxy was relatively inert in acetone, softened in alcohol after long exposures, could not be chemically etched which reduced the conductivity of the epoxy, but could be electro-chemically

* Epoxy Technology Inc., P.O. Box 567, 14 Fortune Drive, Billerica, Mass. 01821.

polished, the epoxy retaining its conductivity.

Various specimen shapes and designs were prepared, see Figure 27 for 3 examples. Methods a) and b) had two or more specimens embedded in the silver epoxy with larger support pieces consisting of a zirconium-chromium-iron alloy at the sides. A slow speed diamond saw was used to cut the specimen into 1 mm thicknesses. They were further thinned by using a grinding jig, lapping them on silicon carbide papers (240, 320, 400 and 600 grit size, respectively) to a thickness of 25 to 50 μm which enabled them to be ready for the final thinning process.

The final thinning was performed by electro-chemical polishing rather than ion-beam thinning as is done with the ceramic materials, because zirconium alloys have not yet been successfully ion-beam thinned (145). The zirconium specimens that have been ion-beam thinned had a mottled surface with much oxide and were not representative of the true microstructure.

Both a) and b) type specimens produced thin foils suitable for TEM observation of the microstructure yet they were difficult to handle. The silver epoxy being very thin, cracked very easily when handled, resulting in a low survival thin foil specimen percentage, after the entire thinning process was completed. To overcome this problem, procedure c) was developed. The 30 mm long by 2.5 mm wide irradiated surface of the specimens were spark machine cut by a thin carbon wafer into 1.2 mm long by 2.5 mm wide pieces. They

were then turned on edge (~ 0.13 mm thick) and stacked together in a carbon block, Figure 27c, where they were glued together with the silver epoxy while being squeezed together by a spring and surrounded by wax, used as a releasing agent. Once set, they were lapped on silica-carbide papers using a grinding jig to a thickness of $50\mu\text{m}$. To avoid handling problems they were glued, using the silver-epoxy to a copper microscope grid whose grid had been cut out with a scalpel to the size of the specimen, Figure 28. The specimens were then electro-chemically thinned in the usual manner, but glyptol (microstop) was used to protect the copper grid as copper is preferentially thinned over zirconium, as zirconium is to the silver epoxy.

CHAPTER 4

Experimental Results

4.1 Irradiation Growth Measurements

The cantilever-beam specimens' deflections, δ and their associated strains are given in Table 9. The deflections are corrected for effects due to the mounting apparatus on the light bench. A multiplying factor of 0.84 is used to change the measured deflections to true deflections of the specimens, since the mirrors are non-centrosymmetric on the mounting apparatus. The corrected deflections are plotted in Figure 29 to show the dependence of the deflections on neon ion dose, specimen composition, and direction of measurement, i.e., texture and grain shape. The average strains, Table 10, which normalize the specimens thickness, using equation 2.1, i.e.,

$$\epsilon = \frac{t \cdot \delta}{l^2}$$

are plotted as a function of dose in Figure 30. The growth strains tend to increase with decreasing alloying addition, i.e., the polycrystalline Zr > Zr-2.5wt%Nb > XL Alloy > Zircaloy-2. From Tables 11 and 12, it can be seen that the growth strains tend to increase with an increase in prism pole texture coefficient orientated in the direction of measurement. The growth strains of the longitudinal specimens, having higher prism pole texture coefficients and elongated grains in the direction of measurement, are always greater than their respective transverse specimens.

4.2 Electron Microscopy

Micrographs of as-received polycrystalline Zr, Zircaloy-2 and Zr-2.5wt%Nb specimens, are shown in Figure 31. Both the Zircaloy-2 and Zr-2.5wt%Nb show a cold-worked structure, having regions of high dislocation density, whereas the polycrystalline Zr specimens are in the annealed condition and show only a few isolated dislocations. Micrographs of irradiated thin foil specimens orientated normal to the irradiated direction are shown in Figures 32-36. The low temperature ($\sim 314\text{K}$) irradiated specimens have a high dislocation density with many small dislocation loops (indicated by arrows). In high contrast conditions ($g \cdot b > 2$, $b = \langle 11\bar{2}0 \rangle$) the irradiation damage appears as extremely dense dislocation tangles, sometimes referred to as having a 'black death' structure. Determining the crystallographic orientation of the thin foil was aided by using reference (146).

Thin foil specimens orientated perpendicular to the neon ion irradiation beam direction are shown in Figure 37 and 38. These thin foils usually had a plateau shape with electro-chemically thinned valleys on one or both sides, as in Figure 37a. The remaining silver epoxy appears as round balls and amorphous material on the surface and to the sides. In Figure 37, the heavily irradiated damage layer appears to be about 60 nm in depth and the entire damage layer is about 100 nm in depth.

The thin foil of the irradiation damage layer shown in Figure 38 has a measured parallax, P of about 2mm. The

thickness of the specimen can be calculated from the equation (65),

$$P = 2TM \sin \Delta\theta/2 \quad (4.1)$$

where M is the magnification, $\Delta\theta$ is the rotation angle and the thickness, T is around 96 nm. The irradiation damage layer depth, d can be calculated, from Figure 39 and the equation:

$$h = T \sin\theta + d \cos\theta \quad (4.2)$$

where h is the observed depth, θ is the rotation angle and d is calculated to be around 51 nm, which gives a rough estimate along with Figure 37 of the heavily irradiation damage layer depth.

In all specimens examined the surface was covered by an oxide layer which in certain contrast conditions appeared as black spots and could be mistaken for the irradiation damage. However, by employing a through-focus series of micrographs the oxide changed in contrast from black to white, Figure 40, thus allowing it to be distinguished from the irradiation damage which remains black. Also, an oxide layer is formed on the cantilever-beam specimen surface during irradiation. The thin foil specimens made normal to this surface had the oxide layer protected during the thinning process and it could be seen on all these specimens, Figure 41. The oxide layers decreased resolution of the irradiation damage by creating Moire fringe patterns and by only allowing thick specimens to be examined. If the oxide layer was on

the bottom side of the thin foil when examined in the electron microscope, the diffraction pattern sometimes had diffraction rings (from the oxide) around the diffracted beams, Figure 41c. The zirconium oxide was indexed as having a mixed monoclinic plus tetragonal structure.

4.3 SAM and X-ray Line Broadening Analysis

The irradiation damage depth distribution should be similar to the neon ion concentration profile. Scanning Auger Microscope (SAM) which can determine surface concentrations was tried in order to detect the neon ion concentration profile by argon sputtering off layers of the specimen surface at a pressure of $\sim 10^{-9}$ torr. The auger-electron-energy spectrum of a cantilever-beam specimen's surface having a dose of 5×10^{20} neon ions/m², after one minute of argon sputtering which corresponds to ~ 10 nm of removed surface is shown in Figure 42, from which zirconium, oxygen, carbon and argon have been positively identified. Neon should have a kinetic energy peak at 800 eV and cannot be seen in Figure 42. Figure 43 shows a concentration profile of monitored zirconium, oxygen and neon kinetic-energy-ranges as a function of argon sputtering time. Only the background noise is indicated by the neon concentration profile. The neon concentration (~ 0.012 at%) may be too low to be detected. The oxygen profile shows the presence of an oxide layer having a thickness of ~ 10 to 20 nm (1 to 2 minutes sputtering time). The oxide layer seen on the

electron microscope thin foils would probably correspond to this thickness.

X-ray line broadening measurements using the Fourier Transform method (147,148) assisted by computer analysis (149,127) of irradiated (5×10^{20} neon ions/m²) and bulk Zircaloy-2 specimen were used to determine the lattice strain and coherent diffraction domain size, and are compared in Table 13. The lattice strain in the irradiated specimen is twice that of the unirradiated specimen. The coherent diffraction domain size is 100 μ m less in the irradiated specimen. These results should be corrected somewhat as the 40KeV K_αCu x-rays penetrate to a depth of 4,000 nm at $\theta = 16^\circ$ and 13,000 nm at $\theta = 55.5^\circ$ (150), whereas the depth of the irradiation damage is only ~100 nm. The lattice strain in the irradiated damage layer of the specimen is expected to be higher and the coherent diffraction domain size to be less than the measured values. The decreased coherent diffraction domain size and increased lattice strain are indications of the increased dislocation density due to irradiation damage plus the addition of the neon ions.

CHAPTER 5

Discussion

As a result of the 100KeV neon ion irradiations, the growth strains vary from 2 to 6×10^{-5} , depending on composition, texture and grain shape. This growth strain behaviour is in good agreement with previous neutron irradiation data noted previously in the literature review. The growth strains decrease with increasing alloy composition, low prism pole texture coefficients (or high basal pole texture coefficient) and short grains in the direction of growth measurement.

The Holt-Ibrahim model can be used to analyse this data. The growth and creep anisotropy factors must be determined before the irradiation growth and creep rates can be calculated. The Holt-Ibrahim model assumes creation of equal numbers of vacancies and interstitials during the irradiation damage event and partitioning of these defects to various sinks. The growth anisotropy factor was reasoned to be a function of the vacancy mobility to grain boundaries and the free surface, as well as, a function of texture. Table 14 lists the thermal concentration and jump frequency of interstitials and vacancies. It can be seen that the interstitials are always quite mobile and the vacancies change from immobile at 300K to mobile at the higher temperatures. The irradiation experiments were performed at an average temperature of 314K. At a migration energy of 1.35eV,

vacancies would be considered immobile and thus at this temperature the Holt-Ibrahim model, which relies on the mobility of vacancies could not be used. However, Hood (150) has shown that rapid diffusion of point defects can occur along dislocations and grain boundaries. The activation energy for these regions is about a half of that for a low dislocation density region, i.e., $0.5 Q_{\text{mobility}}$. MacEwan and Carpenter (36) and Holt (28) speculate that vacancies can be 'short circuited' to grain boundaries or free surfaces. Buckley and Manthorpe (65) have calculated a vacancy mobility of 0.65eV in Zr-2.5wt%Nb during 1MeV Electron Irradiation studies. Using this value the vacancies become 'sluggishly' mobile at 300K. The electron micrographs presented in this thesis show an extremely high dislocation density and thus, the Holt-Ibrahim model may be applicable to model the data at 314K. The trapping of point defects by alloying additions has been shown to be very significant with the XL alloy and Zircaloy-2, both having the highest alloying additions and the least growth strains. The polycrystalline Zr specimens, being unalloyed, have the largest growth strains. The trapping of point defects by impurities is as yet unknown. It is speculated that in zirconium there is impurity segregation which is playing a part in point defect clustering, perhaps by altering the long-range strain fields of dislocations (152).

If the vacancies are so sluggishly mobile that they do not contribute to the measured growth strain, then an

alternative explanation for the measured growth strain may be found within the irradiation damage defect structure. At low temperature irradiations, small dislocation loops (<2nm) have been seen to form (68). The cores of the dislocation loops, whether they be interstitial or vacancy in character contribute to a volume strain, $\Delta V/V$ given by (16) as:

$$\frac{\Delta V}{V} = 2\pi r_o b c N \quad (5.1)$$

where, r_o is the radius of the zirconium atom, (0.178nm), b is the Burger's vector ($b=0.32\text{nm}$), c is the radius of the loop and N is number density of dislocation loops. From the electron micrographs, the dislocation density is seen to be so high as to form a 'black death' type of structure. From the increase in lattice strain, ϵ and decrease in coherent diffraction domain size, D an increase in dislocation density can be calculated by (153):

$$\rho_D = 3n/D^2 \quad (5.2)$$

and,

$$\rho_\epsilon = K(\epsilon^2)/\underline{b}^2 \quad (5.3)$$

where ρ_D is the dislocation density calculated from domain size, ρ_ϵ is the dislocation density calculated from the lattice strain, n and K are geometric factors, $\underline{b}$ is the Burger's vector (154). The value of K is taken as 10 from the work of Aqua and Owens (135). This assumes that the dislocation strain fields have a spatial distribution between Cauchy and Gaussian and that the strain fields extend about 50 nm.

The value of n can be taken as 1 after Williamson and Smallman (156). The dislocation densities calculated for the irradiated and bulk Zircaloy-2 material is given in Table 15. When $\rho_e < \rho_D$ a polygonized cell structure is supposed to exist (153). This is seen in the electron micrographs of the irradiated and bulk Zircaloy-2 specimens, Figures 31b, 35 and 36. The calculated dislocation density from the lattice strain in the irradiated material is greater than a half order in magnitude than the bulk specimen. Due to the x-ray depth penetration being much greater than the irradiation damage depth the measured strain, $\Delta d_{hkl}/d_{hkl}$ in the damaged layer is expected to be greater and the dislocation density could approach 10^{15} lines/m³.

Low temperature irradiations (<550K) produce dislocation loops with a diameter <2nm (68,70). Carpenter (15) states that a dislocation density of $\sim 10^{15} \text{ m}^{-2}$ corresponds to dislocation loops, 2 nm in diameter at a density of $\sim 2 \times 10^{23} \text{ m}^{-3}$, as the saturated loop density in neutron irradiated zirconium single crystals, which is an order of magnitude greater than that found by Northwood et al, Figure 8.

If these values are used in equation 5.1 a volume strain of 7.16×10^{-5} is obtained, which is greater than the strain measured by the deflection of the cantilever-beam specimens. In a polycrystalline material the strain is expected to be texture dependent. The growth strains measured from the deflection of the Zircaloy-2 irradiated cantilever-beams and the texture-dependent volume strains, i.e., $(\Delta V/V) \times P_d$, calculated from equation 5.1, are compared in

Table 16 and, as can be seen, the agreement is good.

The growth strain contribution due to dislocation loop alignment, which is texture dependent is given by (93):

$$\epsilon = \frac{\pi b N d^2}{8} \quad (5.4)$$

and has been used in many growth models (47,93,99), but both interstitial and vacancy loops exist at high temperatures (5) and are expected to exist at low temperatures as well (36). The two types would cancel out their growth strain contributions.

Fainstein and Pedraza (127) state that there are three temperature regions in which different mechanisms must dominate. Their model states that the high temperature range is $>535\text{K}$, yet the experimental data by Adamson et al (10), Figures 3 and 7 shows a change in growth mechanism at $\sim 873\text{K}$. At this temperature little or no irradiation damage has been observed by electron microscopy (34). At lower temperatures, in the intermediate range $\sim 600\text{K}$, large dislocation loops are formed and the pre-existing dislocation structure decreases in density due to irradiation enhanced recovery (5). Very little electron microscopy has been performed on the low-temperature ($<350\text{K}$) irradiate specimen due to the difficulty associated in characterizing the nature of the irradiation damage (68). Also, if the vacancy mobility is extremely low, the grain boundaries and free surface would not be expected to contribute to the growth strain and the growth strain measured would be expected to be similar to single crystal

growth data. The comparison between the single crystal growth data of Carpenter et al (15), Figure 1 with the measured growth strains of the deflected cantilever-beam specimens, Figure 31, is good. Both having growth strains of $\sim 10^{-4}$ for zone refined zirconium (which includes the polycrystalline zirconium specimens) at temperatures $\leq 350\text{K}$. The saturation in growth strain could be due to a saturation of vacancies and interstitials within the lattice, enabling the recombination rate to become equal to the point defect creation rate. Solute additions such as Sn and Nb can trap point defects (65,66,157) resulting in an earlier saturation in growth strain for alloyed material. In this model, irradiation enhanced creep would not be expected to play a role in the saturation of the deflection of the cantilever-beam specimens, recombination of point defects being the primary mechanism.

Two experimental observations support the mobility of vacancies model. First, on the rare occasion after an irradiation deflection measurement which was taken immediately after neon ion irradiation, the specimen would continue to bend and then, reverse its deflection so that ultimately the deflection was around the original deflection measurement. This observation could be due to vacancies moving sluggishly to the surface, creating a measurable growth strain (increase in deflection). The increased stress on the damage layer could result in creep of the damage layer back to its original shape. The second observation supporting the mobility of

vacancies deals with the oxide layer formed during irradiation. The oxide layer is formed both during neon ion irradiation and, after neon ion irradiation during x-ray irradiation. No zirconium specimen to date has been reported to oxidize by x-ray irradiation only. The initial zirconia thickness ranges from 1 to 5 nm (158). After irradiation to a neon ion dose of 5×10^{20} neon ions/m², the zirconia thickness has been measured by SAM to be ~15 nm. The growth of zirconia has been characterized to occur by an oxygen exchange mechanism in which the oxygen ions diffuse from the atmosphere/zirconia surface to the zirconia/metal surface (159,160). It is believed that charged vacancies move from the metal/zirconia surface to the zirconia/atmosphere surface (160). The mobile vacancies can annihilate at the metal/zirconia surface and enhance the growth of oxide by supplying an excess of vacancies to move along the vacancy gradient to the zirconia/atmosphere surface. X-rays are known to dissipate charges. The vacancies exist as a negative charge and their mobility would be enhanced by x-ray bombardment and charge dissipation by reacting with the oxygen in the air.

This master's thesis is only part of an ongoing project which still requires much experimental work and theoretical reasoning in order to contribute to the understanding of irradiation growth and perhaps, to irradiation enhanced creep, in order to predict the irradiation behaviour in currently used, as well as, future planned zirconium alloys

for the CANDU nuclear generating system and others. The significance of this work cannot be over-emphasized as the lifetime of the nuclear core material will be limited by dimensional changes within the reactor core rather than from material failure such as creep rupture (161).

CHAPTER 6

Conclusions

1) The growth behaviour of 100keV neon ion irradiated cantilever-beam specimens agrees well with that published in the literature for neutron irradiated materials.

2) Irradiation growth increases with decreasing alloying additions, with an increase in prism pole texture coefficient and elongated grains in the direction of measurement.

3) The irradiation damage layer, as seen by TEM is about 100 nm in depth, with a heavily damaged layer around 60 nm in depth.

4) From the literature review some general conclusions can be made:

- a) Annealed polycrystalline zirconium has a saturation growth strain an order of magnitude greater than that of single crystal data. Thus irradiation growth due to the presence of grain boundaries is significant.
- b) Cold-worked and cold-worked-plus-stress-relieved zirconium does not have a saturation growth strain but continues at a steady-state-rate proportional to the degree of cold work. Thus the irradiation growth rate is proportional to the pre-irradiation dislocation density.
- c) Irradiation growth decreases with an increase in alloying additions to the alpha zirconium phase.

- d) Impurities increase the initial growth transient of irradiated single crystals but do not decrease the saturating growth strain.
- e) There appears to be three temperature ranges in which different, or a combination of growth mechanisms may be occurring. At high temperatures ($T > 800\text{K}$), few or no irradiation defects and dislocation are seen by TEM and the primary growth mechanism is thought to be controlled by diffusion of point defects, biased to their respective sinks. At intermediate temperatures (400K to 800K) could be due to the irradiation produced defects seen by TEM and diffusion of point defects to their respective sinks. At low irradiation temperatures ($< 400\text{K}$) more work still needs to be performed to identify the irradiation growth mechanism.

CHAPTER 7

Suggestion for Future Work

7.1 Improvements to Experimental Procedure

First of all, I would like to suggest two ways of improving the basic experimental procedure. Since a laser is already used to measure the deflections of all the cantilever-beam specimens on the optical bench, it can also be used to give an estimate of the thickness of the specimens, and be used to measure the variation of the specimens thickness along its length, ($\Delta \epsilon = \epsilon t^{-1} \Delta t$). This can be easily accomplished by placing the specimen onto a jig having a fixed position in line with the laser beam. The specimen could be held onto the jig by wax, under pressure ensuring a flat bond. A razor blade can be fixed so as to only allow laser light to pass between it and the specimen, the razor blade being mobile and set as a reference against the surface of the jig, Figure 44. The razor blade can be brought flush against the specimen's surface by a micrometer, measuring the thickest part of the specimen and any deviations from this thickness will result in a lines diffraction pattern on the wall behind the specimen, due to the open slit between the specimen and razor blade. The width between the diffraction lines is inversely proportional to the slit width.

The other basic improvement would be used to reduce the oxidation and contamination of the specimen's surface during irradiation. An electric field builds up on the specimen's surface due to the impingement of positively charged

ions during irradiation, as well as, from the escape of secondary electrons due to the penetration of the bombarding ions. According to Fourie (163) this electric field enhances diffusion of hydrocarbons, and it is thought for zirconium, oxygen as well. Fourie (163) suggests neutralizing the positive charged produced per unit time by using a low energy electron gun, positioned in the specimen chamber (there is sufficient space), so that a beam of low energy electrons could be allowed to impinge directly on the whole area which is being bombarded by the high energy ion beam. Experimental evidence shows that the rate of contamination in the 'field absent' conditions is orders of magnitude less than in the 'field present' condition.

7.2 Improved Thin Foil TEM Specimen Preparation

The thin foil specimens made perpendicular to the neon ion beam direction were very brittle and hard to handle which was overcome by fastening the specimens onto microscope grids. Only a few of these specimens were actually made into thin foils by electro-chemical thinning as an intermediate thinning step was thought to be lacking (a dozen specimens are still left in this stage). These lamellar type specimens, being around 0.001 to 0.002 inches thick were still a bit too thick for making good thin foil specimens of the irradiation damage layer. The specimen should have been ion beam thinned until a hole just perforated the specimen. The ion beam would thin the zirconium and silver-epoxy at approximately the same rate, leaving many individual irradiation damage

layer specimens close to perforation. The structure left behind by the ion beam thinning would then be removed by a final electrochemical thinning, producing a TEM thin foil specimen. The alternative method by nickel deposition, Figure 45, should also be continued as a method to protect the zirconium specimens surface.

7.3 Future Growth Experiments

There are still a couple of experiments that are required in order to help determine the growth mechanism of the cantilever-beam specimens. The first, irradiating the specimen at a higher temperature ($\sim 575\text{K}$) was begun last year and has recently been accomplished successfully (125). The second is an annealing experiment in which the deflection of the cantilever-beam should be monitored as a function of annealing time. For the Zircaloy-2 specimen, annealing at a temperature of 675 K for 20 hours is required for the number of atom sites involved in the irradiation damage to decrease, yet, the volume strain calculated by equation 5.1 is a function of loop radius and the cantilever-beam deflection would be expected to change before 20 hours as the dislocation loops should grow at the expense of others (5). Both of these experiments should give a better idea as to the roles of the irradiation damage, as well as, the vacancies and their mobility in order to help understand both of their particular contributions to the measured growth strains.

7.4 Analysis of Irradiation Damage

The low temperature irradiation damage seen in the electron micrographs is very hard to characterize. Cann and others (6,8,70,71), have computer modelled what the low temperature irradiation damage should look like under specified conditions. Woo (68,164) has taken electron micrographs of low temperature irradiation damage in the condition specified by computer simulation of hcp titanium material. Along with Carpenter, their first analysis of the damage was to characterize it as interstitial loops, but this was doubted on the basis that most low temperature, heavy ion irradiation damage reported has been vacancy in character (165,166) and light ion irradiation damage has been interstitial in character (165). Thus the difficulty in characterizing the irradiation damage is a problem and it is recommended that this type of work be supported in order to help determine the role of low temperature irradiation damage during irradiation growth.

7.5 High Voltage Electron Microscopy

It is recommended that the role of alloying additions and impurities during irradiation growth be studied by High Voltage Electron Microscopy and Electron Energy Microanalysis so as to determine the partitioning of vacancies and interstitials to sinks. This can be studied by measuring the change in an ordered alloy and the segregation of impurities during in-situ irradiation.

7.6 Cantilever-Beam Irradiation Growth Experiments

It is recommended that a series of cantilever-beam irradiation growth measurements should be performed with specimens characterized by TEM and x-ray analysis both before, during and after irradiation. The specimen should be made from flat plate material having a thickness greater than 3 mm so as to allow making specimens with an irradiated surface in the longitudinal normal, transverse normal and radial normal directions. The specimen's condition could be varied by cold working (dislocation density and grain shape), varying alloying additions, by heat treating (dislocation density, grain size, segregation of alloying elements, and phases present), and the irradiation condition by varying the temperature of irradiation. After each change in the specimen's condition a new growth and creep anisotropy factor should be determined. As well, the method for determining the growth and creep anisotropy factors given in this thesis should be tested for their validity.

7.7 Other Possible Areas of Research

Two areas of research that have as yet not been fully considered for irradiation growth and creep are non-Hookian contractions around dislocation cores and the significance of the shock wave from the deposited energy during nuclear stopping of energetic projectiles.

Table 1
Key to Figure 3

Symbol	Temperature of Irradiation (K)	Texture Coefficient	Material	Ref.
●	353	---	Zircaloy-2	17
▼	555	0.122	Zircaloy-4	18
⊙	339	0.082	Zircaloy-4	18
□	616	0.14	Zircaloy-4	18
◇	573	---	Zircaloy	19
◆	573	---	Zircaloy	20
△	573	---	Zircaloy	21
■	573	---	Zircaloy	22
▽	553	0.10	Zircaloy-2	23
■	353	0.07	Zircaloy-2	23
●	353	0.10	Zircaloy-2	23
☆	353	0.16	Polycrystalline Zirconium	23
⊛	673	0.21	Zircaloy-4	10
★	653	0.12	Zircaloy-4	10
▲	573	0.07	Zircaloy-2	24
▽	523	0.12	Zircaloy-4	25
▲	570	---	Zircaloy-4	26
▽	570	---	Zircaloy-2	26
△	570	---	Zircaloy-4	26

Table 2

Experimental Values for Cold-Worked Materials of the Fluence Index q in the
Equation $\epsilon = \text{constant} \times (\phi t)^q$

Alloy	Metallurgical Condition	Fluence Index q	Reference
Zircaloy-4	78% CW + 4h/780 K	0.93	(18)
Zircaloy-2	78% CW + 770K SR (PWR fuel cladding)	0.84	(22)
Zircaloy-2	70% CW + 770K SR (PWR fuel cladding)	0.89	(21)
Zircaloy-2	70% CW + 770K SR (PWR fuel cladding)	0.50	(20)
Zircaloy-2	70% CW + 770K SR (SGHWR fuel cladding)	0.60	(29)
Zircaloy-2	CW + SR (fuel cladding)	0.50	(30)
Zircaloy-2	cold-worked (pressure tube material)	0.70	(31,17)
Zircaloy-2	cold-worked (SGHWR fuel cladding)	1.0	(5)
Zircaloy-2	20% CW (pressure tube material)	0.56	(10)
Zircaloy-2	20% CW (pressure tube material)	0.34	(10)
Zircaloy-2	25% CW (plate at 353K)	0.28	(23)
Zircaloy-2	25% CW (plate at 553K)	0.63	(23)

Table 3
Characterization of Damage in Irradiated Zirconium Alloys

Alloy	Type of Irradiation	Temperature (K)	Fluence	Characterization of Damage		Reference
				Burger's Vector	Vacancy (V)/Interstitial (I) Ratio	
Zirconium	Neutron	473	$8 \times 10^{23} \text{ m}^{-2}$	$a/3\langle 11\bar{2}0 \rangle$	---	15,44
Zirconium	Neutron	523	$1.8 \times 10^{24} \text{ m}^{-2}$	$a/3\langle 11\bar{2}0 \rangle$	55%V/45%I	15,44
Zirconium	Kr ⁺ Ions	R.T.	$10^{17} \text{ ions m}^{-2}$	$a/3\langle 11\bar{2}0 \rangle$	100%I	7
Zirconium	Neutron	473 & 673	$1 \times 10^{24} \text{ m}^{-2}$	$a/3\langle 11\bar{2}0 \rangle$	---	45
Zirconium	Neutron	673	$7.4 \times 10^{24} \text{ m}^{-2}$	$a/3\langle 11\bar{2}0 \rangle$	---	47
Zirconium	1MeV e ⁻	573-873	4.8 dpa	$a/3\langle 11\bar{2}0 \rangle$ plus 5% non-basal	---	9
Zirconium	Neutron	R.T.	$1.4 \times 10^{24} \text{ m}^{-2}$ (+ anneal)	$a/3\langle 11\bar{2}0 \rangle$	75%V/33%I	39
Zirconium	Neutron	573	$3 \times 10^{23} \text{ m}^{-2}$	$a/3\langle 11\bar{2}0 \rangle$	---	39
Zirconium	Neutron	573	$8 \times 10^{23} \text{ m}^{-2}$	$a/3\langle 11\bar{2}0 \rangle$	---	47
Zirconium	Neutron	573-673	$7 \times 10^{23} \text{ m}^{-2}$	$a/3\langle 11\bar{2}0 \rangle$	Both present	48
Zirconium	100KeVHe ⁺	623-773	5 dpa	$a/3\langle 11\bar{2}0 \rangle$	---	49
Zirconium	Neutron	675	$6.4 \times 10^{23} \text{ m}^{-2}$	$a/3\langle 11\bar{2}0 \rangle$	85%V/15%I	50
Zirconium	Neutron	510-723	$1.8 \times 10^{24} \text{ m}^{-2}$	$a/3\langle 11\bar{2}0 \rangle$	Both present	51
Zirconium	Neutron	573-673	$1 \times 10^{25} \text{ m}^{-2}$	$a/3\langle 11\bar{2}0 \rangle$	Excess I at 573K Excess V at 673K	52
Zirconium	Neutron	670	$1.1 \times 10^{25} \text{ m}^{-2}$	$a/3\langle 11\bar{2}0 \rangle$	59%V/41%I	53
Zirconium	Neutron	673	$1 \times 10^{25} \text{ m}^{-2}$	$a/3\langle 11\bar{2}0 \rangle$	70%V/30%I	53, 9
Zirconium	Neutron	573	$1.1 \times 10^{25} \text{ m}^{-2}$	$a/3\langle 11\bar{2}0 \rangle$	26%V/74%I	53

Table 3 (Continued)

Alloy	Type of Irradiation	Temperature (K)	Fluence	Characterization of Damage		Reference
				Burger's Vector	Vacancy (V)/Interstitial (I) Ratio	
Zirconium	Neutron	673	$4 \times 10^{25} \text{ m}^{-2}$	$a/3\langle 11\bar{2}0 \rangle$	---	54
Zirconium	Neutron	673	$1 \times 10^{25} \text{ m}^{-2}$	$a/3\langle 11\bar{2}0 \rangle$	66%V/34%I	55
Zirconium	Neutron	700	$6.3 \times 10^{23} \text{ m}^{-2}$	$a/3\langle 11\bar{2}0 \rangle$	---	56
Zirconium	Neutron	670	$3.3 \times 10^{23} \text{ m}^{-2}$	$a/3\langle 11\bar{2}0 \rangle$	77-67%V/ 23-33%I	56
Zirconium	Neutron	670	$6.4 \times 10^{23} \text{ m}^{-2}$	$a/3\langle 11\bar{2}0 \rangle$	91-67%V/ 9-33%I	56
Zirconium	Neutron	670	$1.8 \times 10^{24} \text{ m}^{-2}$	$a/3\langle 11\bar{2}0 \rangle$	63-45%V/ 37-55%I	56
Zirconium	Neutron	623	$5.3 \times 10^{23} \text{ m}^{-2}$	$a/3\langle 11\bar{2}0 \rangle$	53-37%V/ 47-63%I	56
Zirconium	1MeV e'	773	1 dpa	$a/3\langle 11\bar{2}0 \rangle$	Suggested I	5,57
Zirconium	Neutron	773	$1.8 \times 10^{24} \text{ m}^{-2}$	$a/3\langle 11\bar{2}0 \rangle$ $a/6\langle 20\bar{2}3 \rangle$	Both Present 100%V	64
Zirconium	Neutron	573	0.8 dpa	$a/3\langle 11\bar{2}0 \rangle$	---	67
Zirconium	Neutron	673	0.68 dpa	$a/3\langle 11\bar{2}0 \rangle$	64%V/53%I	67
Zirconium	Neutron	773	0.57 dpa	$a/3\langle 11\bar{2}0 \rangle$	Tangles Only	67
Zirconium	Neutron	923	0.4 dpa	$a/3\langle 11\bar{2}0 \rangle$	No Damage	67
Zirconium	HVEM	673	---	$a/3\langle 11\bar{2}0 \rangle$ $+a/6\langle 20\bar{2}3 \rangle$ $+a/3\langle 1\bar{2}13 \rangle$	Both Present 100%V 100%V	63

Table 3 (Continued)

Alloy	Type of Irradiation	Temperature (K)	Fluence	Characterization of Damage		Reference
				Burger's Vector	Vacancy (V)/Interstitial (I) Ratio	
Zirconium	HVEM	773	6.3×10^{-3} dpa	$a/3\langle 11\bar{2}0 \rangle$	Assumed 100% I	65
Zirconium	HVEM	315-725	15 dpa	$a/3\langle 11\bar{2}0 \rangle$ + some c component	100% I	41
Zircaloy-2	5MeV Ni ⁺⁺	573-873	7-87 dpa	$a/3\langle 11\bar{2}0 \rangle$ + some c component	---	58,59
Zircaloy-2	Neutron	563-600	$0.7-2.5 \times 10^{25} \text{ m}^{-2}$	$a/3\langle 11\bar{2}0 \rangle$ + some c component	---	59
Zircaloy-2	Neutron	523-610	$0.15-2.5 \times 10^{25} \text{ m}^{-2}$	$a/3\langle 11\bar{2}0 \rangle$ + some c component	---	60
Zircaloy-2	1MeV e ⁺	573-873	4.8 dpa	$a/3\langle 11\bar{2}0 \rangle$ + 50% non basal	---	46
Zircaloy-2	Neutron	573-673	$1 \times 10^{24} \text{ m}^{-2}$	$a/3\langle 11\bar{2}0 \rangle$ + $a/3\langle 11\bar{2}3 \rangle$	---	45
Zircaloy-2	Neutron	573	$1.5 \times 10^{24} \text{ m}^{-2}$	$a/3\langle 11\bar{2}0 \rangle$	---	61
Zircaloy-2	Neutron	573	$4 \times 10^{23} \text{ to } 1 \times 10^{26} \text{ m}^{-2}$	$a/3\langle 11\bar{2}0 \rangle$	---	37
Zircaloy-2	Neutron	873	$7.4 \times 10^{24} \text{ m}^{-2}$	$a/3\langle 11\bar{2}0 \rangle$	---	47

Table 3 (Continued)

Alloy	Type of Irradiation	Temperature (K)	Fluence	Characterization of Damage		Reference
				Burger's Vector	Vacancy(V)/Interstitial (I) Ratio	
Zircaloy-2	Neutron	573	$1.5-1.7 \times 10^{24} \text{ m}^{-2}$	$a/3\langle 11\bar{2}0 \rangle$	---	67
Zircaloy-2	Neutron	523	$3.1 \times 10^{24} \text{ m}^{-2}$	$a/3\langle 11\bar{2}0 \rangle$	---	9
Zircaloy-2	Neutron	673	$1 \times 10^{25} \text{ m}^{-2}$	$a/3\langle 11\bar{2}0 \rangle$	55.4%V/44.6%I	9
Zircaloy-2	Neutron	673	$1 \times 10^{25} \text{ m}^{-2}$	$a/3\langle 11\bar{2}0 \rangle$	77 %V/23 %I	55
Zircaloy-2	Neutron	523	$3.1 \times 10^{24} \text{ m}^{-2}$	$a/3\langle 11\bar{2}0 \rangle$	---	55
Zircaloy-2	Neutron	615	$1 \times 10^{25} \text{ m}^{-2}$	$a/3\langle 11\bar{2}0 \rangle$	---	55
Zircaloy-2	1MeV e'	773	1 dpa	$a/3\langle 11\bar{2}0 \rangle$	Suggested 100% I	56
Zircaloy-2	Neutron	573	0.8 dpa	$a/3\langle 11\bar{2}0 \rangle$	---	67
Zircaloy-2	Neutron	673	0.68 dpa	$a/3\langle 11\bar{2}0 \rangle$	63 %V/37 %I	67
Zircaloy-2	Neutron	773	0.57 dpa	$a/3\langle 11\bar{2}0 \rangle$	---	67
Zircaloy-2	Neutron	923	0.4 dpa	---	No Damage	--
Zircaloy-2	HVEM	315-725	23 dpa	$a/3\langle 11\bar{2}0 \rangle$	---	41

Table 3 (Continued)

Alloy	Type of Irradiation	Temperature (K)	Fluence	Characterization of Damage		Reference
				Burger's Vector	Vacancy (V)/Interstitial (I) Ratio	
Zr-2.5wt%Nb	Neutron	573	$3 \times 10^{24} \text{ m}^{-2}$	$\langle a/3\langle 11\bar{2}0 \rangle$	---	47
Zr-2.5wt%Nb	Neutron	553-563	$1.24 \times 10^{24} \text{ m}^{-2}$	$a/3\langle 11\bar{2}0 \rangle$	---	47
Zr-2.5wt%Nb	Neutron	573	0.8 dpa	$a/3\langle 11\bar{2}0 \rangle$	---	67
Zr-2.5wt%Nb	Neutron	673	0.68 dpa	$a/3\langle 11\bar{2}0 \rangle$	---	67
Zr-2.5wt%Nb	Neutron	773	0.57 dpa	---	No Loops	67
Zr-2.5wt%Nb	Neutron	923	0.4 dpa	---	No Damage	67
Zr-2.5wt%Nb	HVEM	773	$6.3 \times 10^{-3} \text{ dpa}$	$a/3\langle 11\bar{2}0 \rangle$	Assumed 100% I	65
Zr-1.15wt%Cr -0.1wt%Fe	Neutron	573	$1.7 \times 10^{24} \text{ m}^{-2}$	$a/3\langle 11\bar{2}0 \rangle$	---	47

Table 4

Effect of Alloy Content on Size and Distribution
of Damage in Zirconium Alloys Neutron-Irradiated
at $\sim 300^{\circ}\text{C}$ to a Fluence of $\sim 1.5 \times 10^{24} \text{ n/m}^2$ ($E > 1 \text{ MeV}$)

Alloy	Alloying Element in Solid Solution	Damage Mean Size (nm)	Density (m^{-3})
Zirconium	0	18	0.6×10^{-21}
Zr-1.15wt%Cr -0.1wt%Fe	<0.2wt%Cr	13	1.0×10^{22}
Zr-2.5wt%Nb	$\sim 0.6 \text{ wt\%Nb}$	7	3.7×10^{22}
Zircaloy-2	$\sim 1.5 \text{ wt\%Sn}$	7	3.3×10^{22}

Table 5

Effect of Post-Irradiation Anneal on the Damage Density and
Recovered Strain of a Zircaloy-4 Stress Relaxation
Specimen Irradiated at 570K

Post Irradiation Anneal	Mean Diameter (nm)	Damage Density (m ⁻³)	N _d [*] (m ⁻³)	Strain Recovery (%)
None	6.25	3.58x10 ²²	0.78x10 ²⁵	0
1h/670K	6.8	3.08x10 ²²	0.80x10 ²⁵	Not Measured
5h/670K	8.2	1.91x10 ²²	0.72x10 ²⁵	Not Measured
20h/670K	10.5	1.27x10 ²²	0.78x10 ²⁵	0.02
100h/670K	11.8	0.86x10 ²²	0.67x10 ²⁵	0.04
500h/670K	20.2	0.25x10 ²²	0.57x10 ²⁵	0.07
16h/720K	19.2	0.24x10 ²²	0.5x10 ²⁵	0.08
90h/770K	0	0	0	0.11

Table 6

An Assessment of the Mechanisms That Must Be Considered in a Model for Irradiation Growth

Mechanism	Dimensional Change + expansion - contraction	Thermally Recoverable	Comments on Relative Contribution
Depleted Zones	+	Yes	Could have a large contribution particularly for annealed materials. Detailed Field Ion Microscopy (FIM) required to obtain estimate of their size and density.
Vacancy Loops	-	Yes	Relative contribution would probably depend on irradiation temperature, with greater contribution at higher (~400°C) irradiation temperatures.
Interstitial Loops	+	Yes	Like vacancy loops, relative contribution would depend on irradiation temperature with greater contribution at irradiation temperatures <300°C.
Dislocation Climb	+	No	Negligible contribution to growth of annealed materials, but significant for cold-worked materials.
Dislocation Climb and Glide	+	No	Negligible contribution to growth of annealed materials, but major contribution to growth of cold-worked materials. There is a real need to obtain an estimate of the magnitude of the irradiation induced recovery of pre-existing dislocations in order to better model this particular mechanism.

Continued

Table 6 (Continued)

Mechanism	Dimensional Change + expansion - contraction	Thermally Recoverable	Comments on Relative Contribution
Grain Boundary Absorption	+-	No	The dimensional change is either positive or negative depending on whether either interstitials or vacancies are being absorbed at grain boundaries. The TEM evidence of denuded zones indicates that the absorption of vacancies is the more likely mechanism and that this occurs at higher irradiation temperatures. The relative contribution to the dimensional change is unlikely to be very large.
Hydrides	+	No	Hydride formation causes a volume expansion; however, the effects are likely to be negligible unless there is considerable corrosion and generation of hydrogen.
Residual Stresses	+-	Yes	The effects are likely to be small for annealed materials and greater for cold-worked materials, at least at the early stages of irradiation.
Voids	+	Yes	The contribution from voids is negligible for both annealed and cold-worked materials. Zirconium and its alloys are remarkably resistant to void formation up to fluences of $1 \times 10^{26} \text{ n.m}^{-2}$, unless the material is pre-injected with helium or irradiated in an helium-producing environment (5).

Table 7

Definitions of Parameters Used in Calculations of MacEwan and Carpenter (36)

Symbol	Definition	Value
A_v	Vacancy-interstitial recombination coefficient	30
A_{2v}	Divacancy-interstitial recombination coefficient	45
b	Dislocation Burgers vector (equated with point defect jump distance)	0.32 nm
E_m^i	Interstitial formation enthalpy	0.25 eV
E_f^i	Interstitial formation enthalpy	4.0 eV
E_m^v	Vacancy migration enthalpy	1.3 eV
E_f^v	Vacancy formation enthalpy	1.6 eV
E_m^{2v}	Divacancy migration enthalpy	0.9 eV
f	Fraction of Frenkel pairs surviving instant recombinations	0.5
Q_D	Activation enthalpy for substitutional self-diffusion	2.9 eV
S	Self-diffusion entropy	3.9 K

continued ...

Table 7 (cont'd)

Symbol	Definition	Value
ΔS_f^V	Vacancy formation entropy	1.5 K
ΔS_m^V	Vacancy migration entropy	2.4 K
ν_D	Debye frequency	$5 \times 10^{12} \text{ s}^{-1}$
Ω	Atomic Volume	0.0236 nm^3
ΔV_i	Interstitial relaxation volume	1.5Ω
ΔV_v	Vacancy relaxation volume	-0.25Ω
X_p	Damage production cross-section (excluding instantaneous recombination) ($E > 1 \text{ MeV}$)	$2.3 \times 10^{-25} \text{ m}^2/\text{n}$
E_b^{S-I}	Binding energy between 0.5% solute and interstitial	0.8 eV
E_b^{2V}	Binding energy between two vacancies	0.3 eV

Table 8
Specimen Composition

Material	Sn	Nb	Ni	Cr	Mo	Fe	Si	Ca	Cu	H	N	O
Polycrystalline Zirconium* Sheet	<10	---	<15	<15	20	165	29	<15	<10	2	<10	186
Zircaloy-2**	1.2- 1.7	---	0.03- 0.08	0.05- 0.15	---	0.07- 0.20	--	---	---	<25	<65	900- 1500
Zr-2.5wt%Nb**	---	2.3- 2.7	----	----	---	----	--	---	---	<25	<65	1000- 1400
XL-Alloy**	3.0- 3.5	0.6- 1.0	----	----	0.6- 1.0	----	--	---	---	<25	<65	1000- 1400

* - all values in ppm

** - all values in wt% except H, N, O which are in ppm

Table 9

Specimen Deflections and Strains

Dose ($\text{Ne}^{++}/\text{m}^2 \times 10^{20}$)	Polycrystalline Zr (Longitudinal)			Polycrystalline Zr (Longitudinal)			Polycrystalline Zr (Transverse)			Polycrystalline Zr (Transverse)		
	Meas. $\text{m} \times 10^{-4}$	Corr.* $\text{m} \times 10^{-4}$	Strain** $\times 10^{-4}$	Meas. $\text{m} \times 10^{-4}$	Corr.* $\text{m} \times 10^{-4}$	Strain $\times 10^{-4}$	Meas. $\text{m} \times 10^{-4}$	Corr.* $\text{m} \times 10^{-4}$	Strain $\times 10^{-4}$	Meas. $\text{m} \times 10^{-4}$	Corr.* $\text{m} \times 10^{-4}$	Strain $\times 10^{-4}$
0.5	2.46	2.07	0.25	4.12	3.46	0.42	2.16	1.81	0.24	2.01	1.69	0.22
1.0	3.43	2.88	0.42	4.55	3.82	0.47	2.97	2.49	0.32	32.0	2.69	0.35
2.0	5.26	4.42	0.54	4.83	4.06	0.50	4.50	3.78	0.49	4.50	3.78	0.49
3.25	6.10	5.12	0.63	5.39	4.53	0.55	----	----	----	----	----	----
4.0	----	----	----	----	----	----	5.23	4.39	0.57	5.36	4.50	0.59
5.25	5.33	4.48	0.55	6.17	5.18	0.63	----	----	----	----	----	----
Thickness ($\text{m} \times 10^{-4}$)	1.10			1.10			1.17			1.17		

* corrected values (measured values $\times 0.84$) for measuring geometry.

** strain values, $\epsilon = t.\delta/\ell^2$

Meas. - Measured

Corr. - Corrected

. . . cont'd.

Table 9 (Continued)

Dose ($\text{Ne}^{++}/\text{m}^2 \times 10^{20}$)	Zircaloy-2 (Longitudinal)			Zircaloy-2 (Longitudinal)			Zircaloy-2 (Longitudinal)			Zircaloy-2 (Transverse)		
	Meas. $\text{m} \times 10^{-4}$	Corr. $\text{m} \times 10^{-4}$	Strain $\times 10^{-4}$	Meas. $\text{m} \times 10^{-4}$	Corr. $\text{m} \times 10^{-4}$	Strain $\times 10^{-4}$	Meas. $\text{m} \times 10^{-4}$	Corr. $\text{m} \times 10^{-4}$	Strain $\times 10^{-4}$	Meas. $\text{m} \times 10^{-4}$	Corr. $\text{m} \times 10^{-4}$	Strain $\times 10^{-4}$
0.5	1.19	1.00	0.18	1.22	1.02	0.18	1.19	1.00	0.18	0.74	0.62	0.10
1.0	1.55	1.30	0.23	1.47	1.23	0.22	1.27	1.07	0.19	0.66	0.55	0.09
2.0	1.93	1.62	0.28	1.85	1.55	0.28	1.93	1.62	0.29	1.04	0.87	0.14
4.0	2.21	1.86	0.33	2.16	1.81	0.33	2.16	1.81	0.32	----	----	----
4.6	----	----	----	----	----	----	----	----	----	1.40	1.18	0.19
7.8	2.54	2.13	0.37	2.54	2.13	0.39	2.54	2.13	0.38	----	----	----
Thickness ($\text{m} \times 10^{-4}$)	1.58			1.63			1.60			1.47		

Table 10
Average Specimen Strain

Dose $\text{Ne}^{++}/\text{m}^2 \times 10^{20}$	Zr-2.5wt%Nb (Longitudinal) ($\times 10^{-4}$)	Zr-2.5wt%Nb* (Transverse) ($\times 10^{-4}$)	XL-Alloy* (Longitudinal) ($\times 10^{-4}$)
0.5	0.20	0.04	0.19
1.0	0.27	0.06	1.23
2.0	0.28	0.07	0.28
4.6	0.42**	0.19	0.37
7.8	----	----	0.43

* - one specimen only

** - linear approximation used

Table 9 (Continued)

Dose ($\text{Ne}^{++}/\text{m}^2 \times 10^{20}$)	XL Alloy (Longitudinal)			Zr-2.5wt%Nb (Longitudinal)			Zr-2.5wt%Nb (Longitudinal)			Zr-2.5wt%Nb (Transverse)		
	Meas. $\text{mx}10^{-4}$	Corr. $\text{mx}10^{-4}$	Strain $\times 10^{-4}$	Meas. $\text{mx}10^{-4}$	Corr. $\text{mx}10^{-4}$	Strain $\times 10^{-4}$	Meas. $\text{mx}10^{-4}$	Corr. $\text{mx}10^{-4}$	Strain $\times 10^{-4}$	Meas. $\text{mx}10^{-4}$	Corr. $\text{mx}10^{-4}$	Strain $\times 10^{-4}$
0.5	1.17	0.98	0.19	1.57	1.32	0.20	1.63	1.37	0.19	0.41	0.34	0.04
1.0	1.45	1.22	0.23	1.63	1.37	0.21	2.74	2.30	0.32	0.61	0.51	0.06
1.62	-----	-----	-----	1.78	1.50	0.23	-----	-----	-----	-----	-----	-----
2.0	1.73	1.45	0.28	-----	-----	-----	2.85	2.39	0.33	0.76	0.64	0.07
3.12	-----	-----	-----	3.05	2.56	0.39	-----	-----	-----	-----	-----	-----
4.0	2.29	1.92	0.37	-----	-----	-----	-----	-----	-----	-----	-----	-----
4.6	-----	-----	-----	-----	-----	-----	3.84	3.23	0.45	1.93	1.62	0.19
5.12	-----	-----	-----	3.10	2.67	0.41	-----	-----	-----	-----	-----	-----
7.8	2.67	2.24	0.43	-----	-----	-----	-----	-----	-----	-----	-----	-----
Thickness $\text{mx}10^{-4}$	1.74			1.38			1.26			1.04		

Table 10
Average Specimen Strains

Dose ($\text{Ne}^{++}/\text{m}^2 \times 10^{20}$)	Polycrystalline Zr (Longitudinal) ($\times 10^{-4}$)	Polycrystalline Zr (Transverse) ($\times 10^{-4}$)	Zircaloy-2 (Longitudinal) ($\times 10^{-4}$)	Zircaloy-2* (Transverse) ($\times 10^{-4}$)
0.5	0.34	0.23	0.18	0.10
1.0	0.44	0.34	0.21	0.09
2.0	0.52	0.49	0.28	0.14
3.25	0.59	----	----	----
4.0	----	0.58	0.33	----
4.6	----	----	----	0.19
5.25	0.59	----	----	----
7.8	----	----	0.38	----

* - one specimen only

Table 11

Prism Pole Texture Coefficients

Material	Principal Directions		
	Radial	Transverse	Longitudinal
Zircaloy-2	0.293	0.237	0.475
Zr-2.5wt%Nb	0.301	0.269	0.430
XL Alloy	0.378	0.159	0.463
Polycrystalline Zr	-----	-----	-----

Table 12

Grain Shape

Material	Grain Length in Principal Directions		
	Radial	Transverse	Longitudinal
Zircaloy-2	8.46	10.21	12.60
Zr-2.5wt%Nb	0.50	2.00	14.00
XL Alloy	-----	-----	-----
Polycrystalline Zr	-----	-----	-----

Table 13

Lattice Strain and Coherent Diffraction Domain Size of Zircaloy-2

Zircaloy-2 Material	Diffraction Planes	Bragg Angle (°)	Line Widths $\frac{B}{A^{-1}}$	Lattice Strain %	Coherent Diffraction Domain Size A°
Irradiated Zircaloy-2	1010	16	0.00193	0.1903	420
	3030	55.5	0.00725		
Bulk Zircaloy-2	1010	16	---	0.0923*	520*
	3030	55.5	---		

* Ref. (126)

Table 14

Thermal Concentration and Jump Frequency of Interstitials and Vacancies (28)

	Temperature Range			Boltzman Equation*
	300K	600K	900K	
Interstitial Concentration (m ⁻³)	6.7x10 ⁻⁶⁷	2.6x10 ⁻³³	4x10 ⁻²²	10exp(-4/kT)
Interstitial Jump Frequency (sec ⁻¹)	1.3 x 10 ⁸	1.2x10 ¹¹	1.1x10 ¹²	1x10 ¹⁴ exp($\frac{-0.35}{kT}$)
Vacancy Concentration (m ⁻³)	1.2x10 ⁻²³	7 x10 ⁻¹²	5.8x10 ⁻⁸	4exp($\frac{-1.4}{kT}$)
Vacancy Jump Frequency (sec ⁻¹)	8.5x10 ⁻⁹	1.8x10 ³	1.1x10 ⁷	4x10 ¹⁴ exp($\frac{-1.35}{kT}$)
Dislocation Enhanced Vacancy Jump Frequency (sec ⁻¹)	4.9x10 ³	1.4x10 ⁹	9.2x10 ¹⁰	4x10 ¹⁴ exp($\frac{-0.65}{kT}$) **

* - Boltzman Constant, k = 8.62x10⁻⁵ eV k⁻¹

** - reference (65)

Table 15

Dislocation Densities Calculated by X-Ray Analysis

Zircaloy-2 Material	Lattice Strain %	Coherent Diffraction Domain Size, A°	ρ_{ϵ} (m ⁻²)	ρ_D (m ⁻²)
Irradiated	0.1903	420	3.54×10^{14}	5.67×10^{14}
Bulk	0.0923	520	0.832×10^{14}	3.70×10^{14}

Table 16

Comparison of Volume and Growth Strains

Zircaloy-2 Material	Texture Coefficient P_d	Volume Strain ($\Delta v/v$) $\cdot P_d$	Growth Strain ϵ
Longitudinal	0.430	3.08×10^{-5}	3.5×10^{-5}
Transverse	0.237	1.70×10^{-5}	1.9×10^{-5}

Figure 1 Axial growth strains for single crystals of zirconium as a function of Neutron Fluence and Temperature.

Figure 2 Axial growth strains for single crystals of zirconium as a function of Neutron Fluence and Purity.

Figure 3 Summary of the irradiation growth data for annealed polycrystalline zirconium, zircaloy-2 and zircaloy-4.

Figure 4 Irradiation growth behaviour of cold-worked or cold-worked plus stress relieved zircaloy.

Figure 5 Irradiation growth in 25% cold-worked zircaloy-2 at 353K and 553K.

Figure 6 Growth transients during irradiation temperature cycling of 25% cold-worked zircaloy-2.

Figure 7 Irradiation growth of annealed zircaloy as a function of irradiation temperature.

Figure 8 Damage accumulation in annealed zircaloy-2 at about 573K. N is the density of the visible defects; $\bar{d}$ is the mean diameter of the defects; and N_d is the number of atom sites involved in the damage, assuming that all defects are perfect loops lying on prism planes.

$z = 11\bar{2}3$

$10\bar{1}0$

$11\bar{2}3$

$11\bar{2}3$

Figure 9 Corduroy contrast seen in thin foils of neutron irradiated zircaloy-2. All micrographs are the same magnification, a) $z = \langle 11\bar{2}3 \rangle$ b) $z = \langle 10\bar{1}0 \rangle$, c) $z = \langle 11\bar{2}3 \rangle$, d) $z = \langle 11\bar{2}3 \rangle$.

Figure 10 Growth strain as a function of texture coefficient, neutron fluence and temperature.

Figure 11 Comparison of growth in zirconium and its alloys.

Figure 12 Strain recovery during post-irradiation annealing studies at 553K of 25% cold worked zircaloy-2.

Figure 13 Comparison of experimental data for cold-worked zircaloy-2 with theoretical calculations based on different models: I = interstitials, V = vacancies, 2V = divacancies, and SI = solute trapping of interstitials.

Figure 14 Buckley's method of detecting irradiation growth effects produced by irradiating non-fissile metals with fission fragments.

Figure 15 Deflection of zirconium thin foil by irradiating oxide ions.

Figure 16 Deflection of zirconium cantilever-beam by bombarding 100 KeV neon ions.

Figure 17 Profile of 100KeV neon ion irradiation damage in zirconium.

Figure 18 Schematic diagram of mass separator, a similar facility as to the one used to irradiate the cantilever-beam specimens.

Figure 19 Jig used to hold specimen during irradiation.

Figure 20 Cantilever-beam specimen mount and positioning.

Figure 21 Specimens from polycrystalline zirconium rolled sheet.

Figure 22 Specimens from zircaloy-2, XL Alloy and Zr-2.5wt%Nb tube stock.

Figure 23 Mirror for low temperature irradiations consisting of aluminum evaporated onto glass.

Figure 24 Mirror for high temperature irradiations consisting of carbon evaporated on polished high carbon steel.

Figure 25 Apparatus and method used for final thinning of TEM specimen.

Figure 26 Typical thin foil specimen.

a)

b)

c)

Figure 27 Silver epoxy thin foil specimens made perpendicular to the irradiation damaged surface.

Figure 28 Silver epoxy thin foil specimen
attached to a copper microscope
grid.

Figure 29(a) Deflection of irradiated cantilever-beam specimens with neon ion dose showing dependence on specimen orientation.

Figure 29(a) Deflection of irradiated cantilever-beam specimen with neon ion dose showing dependence on specimen orientation.

Figure 29(b) Deflection of irradiated cantilever-beam specimen with neon ion dose showing dependence on specimen composition.

Figure 29(b) Deflection of irradiated cantilever-beam specimen with neon ion dose showing dependence on specimen composition.

Figure 30(a) Cantilever-beam strain as a function of neon ion dose and specimen orientation.

Figure 30(a) Cantilever-beam strain as a function of neon ion dose and specimen orientation.

Figure 30(b) Cantilever-beam strain as a function of neon ion dose and specimen composition.

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Figure 30(b) Cantilever-beam strain as a function of neon ion dose and specimen composition.

(a)

(b)

(c)

Figure 31 TEM micrographs of as-received a) polycrystalline zirconium; b) zircaloy-2 and c) Zr-2.5wt%Nb specimens.

Figure 32 TEM micrographs of irradiated polycrystalline Zr specimens orientated in the 0111 direction showing a high dislocation density and a few larger dislocation loops. Figures b and c are enlargements of areas indicated in Figure a.

Figure 33 TEM micrographs of irradiated polycrystalline Zr specimens orientated in the 1014 direction.

Figure 34 TEM micrograph of irradiated polycrystalline Zr specimen orientated in the 1213 direction.

(a)

(b)

Figure 35 TEM micrographs of irradiation damage in Zircaloy-2 specimens orientated in the a) $5\bar{1}46$ and b) $11\bar{2}0$ direction having a high contrast condition ($g \cdot b > 2$) and a high dislocation density.

Figure 36 TEM micrograph of Zircaloy-2 and enlargement of Figure 35(a) showing a polygonized dislocation structure and a few dislocation loops.

Figure 37 Thin foils of the irradiated surface orientated perpendicular to the neon ion beam direction showing a) plateau shape of thin foil area and b) irradiation damaged depth and distribution.

(a)

(b)

(c)

Figure 38 Thin foil of the irradiated surface orientated perpendicular to the neon ion beam direction where b) and c) are enlargements of a), and c) is rotated 12° with respect to a) and b) which shows a change in damage depth of 20 nm.

Figure 39 Thickness of irradiation damage layer, h when specimen is tilted by an angle θ .

Figure 40 Oxide formed on thin foil surface during thinning process showing over and under focussing contrast conditions.

(a)

(b)

(c)

Figure 41 Oxide formed during neon ion irradiation protected by thin foil thinning process where b) is zirconium metal thinned to the oxide layer and c) shows diffraction oxide rings around diffracted beams having a 1014 diffraction pattern.

Figure 42 Auger electron kinetic energy spectrum of zircaloy-2 specimen irradiated by 5×10^{20} neon ions/m², after sputtering the surface by argon ions for one minute.

Figure 43 Profile of zirconium and oxygen composition as a function of sputter time (surface depth).

Figure 44 A method which could be used to measure the specimen's thickness deviations.

Figure 45 Nickel deposition on a zirconium surface.

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Appendix A

Neon Ion Beam - Zirconium Target Interactions

In a simplified physical description, at 100 KeV the accelerated neon ions penetrate the Zirconium target where all the electrons of the neon ion are stripped off and the bare nucleus of charge Ze ($Z_{\text{Ne}} = 10$) proceeds through the medium. The neon ion nucleus suffers coulomb collisions with the target electrons and gradually loses energy. Energy loss to the target nuclei is negligible since they are screened by their electron shells. The projectile proceeds until it is able to capture an electron and so forth until its velocity is smaller than the orbital velocity of the least tightly bound electron. In such a stage of its history it ceases to interact exclusively with the electronic system of the target atoms and the elastic interaction with the target atoms as a whole (via screened coulomb fields) commences. Eventually, the neon ion projectile comes to rest solely due to nuclear collisions (116). The result is a displacement of lattice atoms (displacement spike) and a high generation of localized heat (thermal spike). The displacement of lattice atoms creates a depleted zone commonly referred to as a Seeger Zone (117), Figure A1, which takes into account lattice periodicity, focused momentum transfer, dynamic crowdions which creates spread-out interstitials, and relaxation of the region due to the unstable strain field. Dynamic recovery of the depleted zone in zirconium is expected to

be appreciable since high vacancy concentrations have not been found experimentally (69).

A tremendous number of projectile-target combinations are possible, hence a unified theory of stopping is lacking and always will. Also, it would be unrealistic to expect general theories to completely satisfy all ion-solid systems for all energies of implantation. The Lindhard, Scharff, Schiøtt (LSS) theory (118) of ion ranges and the Winterbon, Sigmund, Sanders theory (119) of damage production offer a very real approximation to the measured quantities.

Schiøtt (120) gives approximations and interpolation rules for ranges and range straggling where he uses a range correction function, $C(A_2/A_1)$ where A_2 is the target mass and A_1 is the ion mass, for low, medium and high energy regions to correct for electronic losses which are important at high mass ratios. The electronic stopping and nuclear stopping contribute additively to the stopping power given by:

$$\frac{d\varepsilon}{d\rho_{\text{tot}}} = \frac{d\varepsilon}{d\rho_{\text{nuclear}}} + \frac{d\varepsilon}{d\rho_{\text{electronic}}} \quad (\text{A.1})$$

where the energy, ε lost per unit path length, ρ is given in reduced units, i.e., KeV and ug/cm^2 , respectively. The equations describing ε and ρ are:

$$\varepsilon = \frac{32.5 A_2}{(A_1 + A_2)^2 Z_1 \cdot Z_2 (Z_1^{2/3} + Z_2^{2/3})^{1/2}} \cdot E \text{ (KeV)} \quad (\text{A.2})$$

$$\rho = \frac{166.8 A_1}{(A_1 + A_2)^2 \cdot (Z_1^{2/3} + Z_2^{2/3})} \cdot R \text{ (ug/cm}^2\text{)} \quad (\text{A.3})$$

where Z_1 and Z_2 are the atomic numbers of the projectile and target, respectively. Using Schitt's range approximations and rules a range of $R_p = 1065A^0$ was obtained for 100 KeV neon ions in zirconium with $\Delta R_p/R_p \sim 1$, for the neon ion beam normal to the irradiated zirconium surface.

If we consider the recoiling atom and the atomic displacements which it produces as the energy is dissipated, a depth profile for the radiation damage can be calculated. To be more precise, the depth profile of energy disposition will have been calculated since the detailed structure of the radiation damage will depend on many parameters, such as the stability of vacancies and interstitials. To simplify the calculation of the spatial distribution of deposited energy, it is customary to assume that electronic energy loss does not contribute to the displacements and that some fraction of the recoil energy is lost in electronic processes. The equations governing the spatial distribution of energy are much like those governing the distribution of ion ranges. There is, however, a major difference between the two distributions. For any single ion path the range distribution shrinks to one point, namely the end point of the ions trajectory. Whereas, the distribution of deposited energy extends over a region taking into account many events whose dimensions are expected to be of the order of the ion range (119). Winterbon (121,3) has calculated the deposited energy profile for 100KeV neon ions penetrating a zirconium target, given in the main text as Figure 17, where the calculations assumed that the accelerated neon ions penetrate

an amorphous zirconium-based material. Channeling occurs only in crystal structures when the ion beam is directed in a crystal's principal direction (119).

The fraction of backscattered particles, i.e., reflection coefficient, γ is dependent on mass, A_2 and charge number of the target atom, Z_2 and ion mass, A_1 , and charge number, Z_1 , and energy, E , of the projectile, given by the equation (116):

$$\gamma = \frac{Z_1^2 \cdot Z_2^2}{E_1^2} \left(\frac{A_1 + A_2}{A_2} \right)^2 \quad (\text{A.4})$$

For 100 KeV neon ions bombarding normal to the zirconium target, $\gamma = 2.37 \times 10^{-5}$. Thus very few neon ions are reflected from the surface. Brown and Davies (122) have measured the sticking factor (inverse of reflection coefficient) of inert gases bombarded at metals and found that ~100% of the bombarded ion species is retained at ion energies greater than 5 KeV. Also, sputtering of the zirconium target material is considered to be low (123), given the experimental conditions of the neon ion irradiation of the zirconium cantilever-beam specimens.

The production of defects in the crystal structure of a neutron or ion-irradiated material begins with a primary knock-on atom and progress through second generation recoiling atoms resulting in a cascade of atomic displacements. If the modified Kinchen-Pease model (124) of this displacement process is used and only elastic collisions are considered, then the average number of Frenkel pairs produced is given by:

$$\nu = 0.8 \frac{E^*}{2E_d} \quad (\text{A.5})$$

where E^* is the amount of energy deposited into atomic collision and E_d is the displacement energy.

In cascades with a high density, the Kinchen-Pease model tends to overestimate the number of Frenkel pairs which survive to alter the physical properties of the irradiated material as the recombination of displaced atoms with vacant lattice sites is neglected.

Figure 1A Schematic representation of radiation damage according to Seeger (117). P denotes the position where the primary knock-on comes to rest.

Appendix B

Expansion of Specimen Due to Neon Ions

The expansion due to the presence of the neon ion, $\Delta V/V$ has been given as follows (125):

Let $\Delta V/V$ = fractional volume change at depth
Z per neon ion, assumed isotropic.

$C(Z)$ = volume concentration of neon ions
in ions/m³.

Assume: $\Delta V/V \propto C(Z)$

and,

$$3 \frac{du}{dz} = A C(Z) \quad (B.1)$$

where A is the volume expansion per neon ion in m³/Ne, u is the elastic displacement vector, and 3 times $\frac{du}{dz}$ since the expansion is considered isotropic. Therefore,

$$\int_0^d 3du = \int_0^d AC(Z) dz \quad (B.2)$$

$$3U_{\max} = A \cdot \phi$$

$$\rightarrow U_{\max} = \frac{1}{3} A\phi \quad (B.3)$$

where $U_{\max}$ is the total, isotropic elastic displacement due to the neon ions. The damage layer of thickness, d would grow by $U_{\max}$ in all 3 directions if unimpeded by the substrate's compressive force, Figure 1B. The corresponding strain increment due to the neon ions is therefore positive and of magnitude,

$$\epsilon_Z^{\text{Ne}} = \frac{1}{3} \frac{A\phi}{d} \quad (B.4)$$

in the Z direction, which is normal to the substrate surface. But this expansion cannot take place in the x and y direction as it is contained by the undamaged substrate.

The unirradiated metal is resisting free expansion of the damaged layer due to volume expansion by a force, F_1 given as:

$$F_1 = E \epsilon_m b(t-d) \quad (B.5)$$

$$= E \epsilon_{d\ell} b(t-d) \quad (B.6)$$

since, $\epsilon_m = \epsilon_{d\ell}$

where b is the width of the cantilever-beam specimen, Figure 1B. Expansion force, F_2 in the unirradiated layer is given by:

$$F_2 = E \epsilon_{d\ell} b d \quad (B.7)$$

The free expansion force of the damaged layer produces a strain $= A\phi/3d$. Therefore if we add the compressive force in equation (B.6) to equation (B.7) we should get the free expansion force,

$$\frac{1}{3} \frac{A\phi}{d} E b d = E \epsilon_{d\ell} b d + E \epsilon_{d\ell} b(t-d) \quad (B.8)$$

Solving for ϵ_d , we get the strain in the x and y direction due to the volume expansion of neon ions as:

$$\epsilon_{x \text{ and } y}^{\text{Ne}} = \frac{1}{3} \frac{A\phi}{t} \quad (B.9)$$

and

$$\epsilon_{x \text{ and } y}^{\text{Ne}} = \frac{1}{3} \frac{A\dot{\phi}}{t} \quad (B.10)$$

which has been used as the strain contribution due to the presence of Neon ions.

Figure B1 Cantilever-beam geometry showing expansion of damage layer if unimpeded by substrate.

Appendix C

Growth and Creep Anisotropy Factors for Neon Ion

Irradiation Growth and Creep in a Cantilever-Beam Specimen

The Holt-Ibrahim model (27) assumes that irradiation growth results from the redistribution of atoms displaced from the lattice by an irradiating species such as bombarding ions. The ions create equal numbers of vacancies, N_v and interstitials, N_i which migrate through the lattice until they recombine or reach a 'y' type sink at which they can precipitate. Thus,

$$\sum_y N_{iy} - \sum_y N_{vy} = 0 \quad (C.1)$$

strain due to the precipitation of point defects at sinks is then given as:

$$\begin{aligned} \frac{\epsilon_d}{\Omega} = & (N_i - N_v) A_d + (N_{iD} - N_{vD}) A_{dD} \\ & + (N_{ig} - N_{vg}) A_{dg} + (N_{is} - N_{vs}) A_{ds} \\ & + (N_{iF} - N_{vF}) A_{dF} \end{aligned} \quad (C.2)$$

where Ω is the atomic volume, ℓ is the dislocation loops, D = dislocations, g = grain boundaries, δ = subgrain boundaries, F = free surface, and A_{dy} is the anisotropy factor for type 'y' sink with respect to the 'd' direction.

The anisotropy factor for dislocations, loops and subgrain boundaries are directly related to crystallographic texture since $a/3\langle 11\bar{2}0 \rangle$ is the dominant Burgers vector and the precipitation of point defects at these sinks will

result in strain in the direction of the Burger's vector.

Thus,

$$A_{dD} = A_d = A_{ds} = \frac{P_d}{2} \quad (C.3)$$

where P_d is the resolved prism pole component in the 'd' direction, Figure 1C.

The direction of strain where a point defect is absorbed at a grain boundary or free surface is the direction of the grain boundary or free surface normal. During forming processes the grains are elongated in the direction of working and compressed in directions normal to the direction of working. The anisotropy factor can be approximated by assuming a simple rectangular grain shape with axes in the three principal directions of strain during working and then,

$$A_{dg} = \frac{1}{d_d} / \left(\frac{1}{d_R} + \frac{1}{d_T} + \frac{1}{d_L} \right) \quad (C.4)$$

where R is the radial direction, T is the transverse direction and L is the longitudinal direction. Similarly, the free surface on the cantilever-beam can be treated as a rectangular grain sliced in half with the annihilation of point defects at the free surface only, Figure 2C, and then:

$$A_{dF} = \frac{1}{d_d} / \left(\frac{1}{d_{\parallel}} + \frac{1}{2d_N} + \frac{1}{d_{\perp}} \right) \quad (C.5)$$

where, $\parallel$ is in the parallel direction to the length of the cantilever beam specimen, $\perp$ is in the perpendicular direction and N is in the normal direction to the free surface, Figure 3C.

Partitioning of the point defects to various sinks

occurs in the absence of applied stress due to the interaction of the strain fields between point defects and sinks. Interstitials are preferentially attracted to dislocations and to loops due to the interaction between the large dilational strain field of interstitials and the long range strain fields of dislocations and loops. Since vacancies have a very much weaker strain field, excess vacancies precipitate at sinks without long range strain fields such as grain boundaries, subgrain boundaries and free surfaces. Thus the total strain due to partitioning of point defects at sinks is:

$$\frac{\epsilon d}{\Omega} = N_{i\ell} A_{d\ell} + N_{iD} A_{dD} - N_{vs} A_{ds} - N_{vg} A_{dg} - N_{vF} A_{dF} \quad (C.6)$$

$$= (N_{i\ell} + N_{iD} - N_{vs}) \frac{P_d}{2} - N_{vg} A_{dg} - N_{vF} A_{dF} \quad (C.7)$$

Since equal numbers of vacancies and interstitials are created by the 100KeV neon ion flux,

$$N_{i\ell} + N_{iD} - N_{vs} - N_{vg} - N_{vF} = 0 \quad (C.8)$$

Thus,

$$\frac{\epsilon d}{\Omega} = (N_{vg} + N_{vF}) \frac{P_d}{2} - N_{vg} A_{dg} - N_{vF} A_{dF} \quad (C.9)$$

Two assumptions can be made. First, the dimensions of the cantilever-beam specimens are $d_{||} = 30 \text{ mm}$, $d_{\perp} = 2.5 \text{ mm}$ and $d_N = 10^{-5} \text{ mm}$ (100nm) which makes the free surface anisotropy factor equal to:

$$A_{dF} = \frac{1}{30} / \left(\frac{1}{30} + \frac{1}{2 \times 10^{-4}} + \frac{1}{2.5} \right)$$

$$\begin{aligned}
 A_{dF} &= 1/d \quad / (1/2d) \\
 &= \frac{2d}{d} \\
 &= \frac{2 \times 10^5}{30} \\
 &= 6.67 \times 10^{-7}
 \end{aligned}$$

which may have negligible contribution to the measured growth strain. Second, since the irradiation damaged layer on the surface of the cantilever beam is much smaller than the grain boundary diameter (10^{-7} m versus 10^{-5} m for small grains), the number of vacancies annihilating at the grain boundaries, N_{vg} should be much less than the number of vacancies annihilating at the free surface, N_{vF} , i.e., $N_{vg} \ll N_{vF}$. The growth strain parallel to the length of the cantilever-beam specimen becomes:

$$\frac{\epsilon_d}{\Omega} = N_{vF} \cdot \frac{P_d}{2} \quad (C.10)$$

Using these two assumptions, the growth anisotropy factor, G_d for measuring the deflection of the cantilever-beam specimen during neon ion irradiation could be only a function of the specimen's surface texture:

$$G_d = \frac{P}{2} \quad (C.11)$$

Also, if these two assumptions were to hold true, the number of vacancies arriving at the free surface could be calculated using experimental data collected by Murgatroyd and Rogerson (26), Table 1C, the growth strain of the cantilever-beam

specimen should be close to their data, if this experiment is to be representative of irradiation growth in zirconium alloys. Calculating the number of vacancies arriving at the free surface after an irradiation dose of 10^{24} n/m^2 yields,

$$\begin{aligned}
 \frac{\epsilon_g}{\Omega} &= K_g G_d \phi \\
 &= K_g N_{vF} \frac{P_d}{2} \phi \\
 N_{vF} &= \frac{\epsilon_g}{\Omega K_g \frac{P_d}{2} \phi} \\
 &= \frac{4 \times 10^{-4}}{2.36 \times 10^{-29} \times 68 \times 10^{-29} \times 10^{24} \times \frac{0.84}{2}} \\
 &= 5.93 \times 10^{28} \text{ m}^{-3}
 \end{aligned}$$

The volume of the cantilever-beam's damage layer can be approximated by:

$$\begin{aligned}
 \text{Specimen Volume} &= \text{irradiated length} \times \text{width} \times \text{depth} \\
 &= 0.03 \times 0.0025 \times 10^{-7} \\
 &= 7.5 \times 10^{-12} \text{ m}^3
 \end{aligned}$$

Thus the net number of vacancies arriving at the free surface is:

$$\begin{aligned}
 N_{vF} &= 5.93 \times 10^{28} \times 7.5 \times 10^{-12} \\
 &= 4.45 \times 10^{17}
 \end{aligned}$$

Thus the first assumption is not valid for reducing equation (C.9) since the net number of vacancies arriving at the free surface is calculated to be very high. The growth anisotropy

factor could then be written as:

$$G_d = \left(\frac{P_d}{2} - A_{dF} \right) \quad (C.12)$$

The second assumption where the net number of vacancies annihilating at grain boundaries is taken to be much less than that annihilating at the free surface, since the grain size is much greater than the damaged layer thickness, is thought to be valid from growth modelling calculations by Bullough (106,127) and Pedraya and Fainstein-Pedraya (128).

Bullough (127) calculated the free surface effect on swelling during heavy-ion irradiation of stainless steel at temperatures $>798K$ using diffusional losses of point defects. In this work he neglected the point defect losses to the bulk material. Later, using the same modelling procedure to determine the sink strength for film surfaces and grain boundaries in zirconium at $773K$, Bullough (106) calculated the error introduced by neglecting the loss of point defects to the bulk material and concluded that only the foil surface sink strength should be used, disregarding the small error ($<5\%$) introduced by ignoring the loss of point defects to the bulk. Also, Pedraya and Fainstein-Pedraya (128) question whether any rate theory approach of the irradiation induced dimensional change phenomena based upon the annihilation of point defects by means of a diffusional process can yield the experimentally measured strain versus dose behaviour at temperatures $<350K$. The growth measurements of the cantilever-beam specimen have been taken at $314K$.

Thus diffusion into the bulk for annihilation of vacancies at grain boundaries would not be probable when using the FPSP (99) growth model. Although the annihilation of vacancies at grain boundaries that intersect the free surface is possible and equation (C.4) could be changed to:

$$\begin{aligned} A'_{dg} &= 1/d_d / (1/d_N + 1/d_{\parallel} + 1/d_{\perp}) \\ &= 1/d_d / (1/d_{\parallel} + 1/d_{\perp}); \quad 1/d_N = \frac{1}{\infty} = 0 \end{aligned} \quad (C.13)$$

and equation (C.9) could be written as:

$$\frac{\varepsilon_d}{\Omega} = (N_{vg} + N_{vF}) \frac{P_d}{2} - N_{vg} A'_{dg} - N_{vF} A_{dF} \quad (C.14)$$

The number of vacancies arriving at the grain boundary is expected to be small as compared to the number of vacancies arriving at the free surface, yet if the bias of vacancies to the free surface is equal to the bias of vacancies to grain boundaries, then the number of vacancies arriving at these two sinks should be equal to their respective areas, B , i.e.,

$$\begin{aligned} \frac{N_{vg}}{B_{vg}} &= \frac{N_{vF}}{B_{vF}} \\ N_{vg} &= \frac{B_{vg}}{B_{vF}} N_{vF} \\ &= KN_{vF} \end{aligned} \quad (C.15)$$

where,

$$B_{vg} = d \times \bar{c}$$

and,

$$\begin{aligned}
 B_{vF} &= \text{free surface area} \\
 &= 30 \text{ mm} \times 2.5 \text{ mm} \\
 &= 75 \text{ mm}^2
 \end{aligned}$$

where d is the damage layer thickness (~ 100 nm) and $\bar{c}$ is the circumference of the grain boundary, i.e.,

$$c = 2\pi r$$

for equiaxed grains, and,

$$c = 2(L + W)$$

for elongated grains, where L is the length and W is the width of the grains. Thus equation (C.14) can be written as:

$$\begin{aligned}
 \frac{\epsilon_d}{\Omega} &= (KN_{vF} + N_{vF}) \frac{P_d}{2} - KN_{vF}A'_{dg} - N_{vF}A_{dF} \\
 &= N_{vF} \left(K \frac{P_d}{2} + \frac{P_d}{2} - KA'_{dg} - A_{dF} \right) \quad (C.16)
 \end{aligned}$$

and the growth anisotropy factor as:

$$G_d = \frac{P_d}{2}(K+1) - KA'_{dg} - A_{dF} \quad (C.17)$$

The deflection, δ of the cantilever-beam due to the growth strain component should be dependent upon texture, grain and specimen shape.

The creep anisotropy factor, C_d depends on texture only and has been given by the Holt-Ibrahim model as:

$$C_d = \epsilon_{Rd} \frac{\sigma_R}{\bar{\sigma}} + \epsilon_{Td} \frac{\sigma_T}{\bar{\sigma}} + \epsilon_{Ld} \frac{\sigma_L}{\bar{\sigma}} \quad (C.12)$$

where $\bar{\sigma}$ and σ_d are the normalized and component stresses in the 'd' direction, ϵ_{dd} are the texture dependent strains in the 'dd' direction, where the first subscript d refers to

the stress direction and the second to the strain direction. The matrix of creep anisotropy factors contains 9 values, only 3 are independent. For the cantilever-beam specimen, let one stress be the reference stress (in our case σ), then the creep anisotropy factor for each direction is:

$$C_N = \frac{\sigma_N}{\sigma_{\parallel}} \epsilon_{NN} + \frac{\sigma_{\perp}}{\sigma_{\parallel}} \epsilon_{\perp N} + \epsilon_{\parallel N} \quad (C.13)$$

$$C_{\perp} = \frac{\sigma_N}{\sigma_{\parallel}} \epsilon_{N\perp} + \frac{\sigma_{\perp}}{\sigma_{\parallel}} \epsilon_{\perp\perp} + \epsilon_{\parallel\perp} \quad (C.14)$$

$$C_{\parallel} = \frac{\sigma_N}{\sigma_{\parallel}} \epsilon_{N\parallel} + \frac{\sigma_{\perp}}{\sigma_{\parallel}} \epsilon_{\perp\parallel} + \epsilon_{\parallel\parallel} \quad (C.15)$$

and, as can be seen from Figure C3, there is a zero stress component in the surface normal direction and a biaxial stress condition is operative for creep during the irradiation of the cantilever-beam specimen. To evaluate the effect of both creep components, the two creep anisotropy factors, $C_{\perp}$ and $C_{\parallel}$ are added () such that the creep anisotropy factor, C_d becomes:

$$C_d = C_{\perp} + C_{\parallel} \quad (C.16)$$

$$= \frac{\sigma_N}{\sigma_{\parallel}} (\epsilon_{N\perp} + \epsilon_{N\parallel}) + \frac{\sigma_{\perp}}{\sigma_{\parallel}} (\epsilon_{\perp\perp} + \epsilon_{\perp\parallel}) + \epsilon_{\parallel\perp} + \epsilon_{\parallel\parallel} \quad (C.17)$$

$$= \frac{\sigma_{\perp}}{\sigma_{\parallel}} (\epsilon_{\perp\perp} + \epsilon_{\perp\parallel}) + \epsilon_{\parallel\perp} + \epsilon_{\parallel\parallel} \quad (C.18)$$

When determining the volume fraction strains, ϵ_{dd} the Holt-Ibrahim model assumes that slip occurs by 80% on prism planes and 20% on basal planes, Figure C4. This assumption is

supported by experimental data by Causey (129) who found the ratios of prism to basal slip are 84/16 in Zr-2.5wt%Nb and 73/27 in Zircaloy-2 at 320K and for both alloys, 65/35 at 570K.

Table C1

Experimental Data by Murgatroyd and Rogerson (26)

Fluence n/m ²	Strain m/m	Texture F _d P _d (1-F _d)		Growth Rate Constant m ² /n	Atomic Volume m ³
10 ²⁴	4x10 ⁻⁴	0.16	0.84	68x10 ⁻²⁹	2.36x10 ⁻²⁹

BASAL POLES, $\Sigma F_d = 1$

PRISM POLES, $\Sigma P_d = 1 + 2\cos 60^\circ = 2$

POLE FRACTION, $1 = F_d + 2P_d$

RESOLVED PRISM
POLE = $\frac{\Sigma P_d}{2}$

Figure C1 Prism and basal poles in h.c.p. structure.

Figure C2 Vacancy concentration profile where a) is equivalent to b) resulting in equation (C.5) (27,111,126).

Figure C3 Defined directions and stress components on irradiated cantilever-beam surface.

Figure C4 Modes of slip in zirconium.

Appendix D

Integration of Equation (2.16)

Integrating the equation

$$\frac{t}{\ell^2} \dot{\delta} = \frac{1}{3} \frac{A}{t} \phi + K_g G_d \phi - K_c C_d \frac{E}{(1-\nu)} t \frac{\delta}{\ell^2} \phi$$

Let:

$$A = t/\ell^2$$

$$B = \frac{1}{3} \frac{A}{t}$$

$$C = K_g G_d$$

$$D = K_c C_d \frac{E}{(1-\nu)} \frac{t}{\ell^2}$$

$$E = B + C$$

$$\begin{aligned} \rightarrow A \dot{\delta} &= B\phi + C\phi - D\delta\phi \\ &= E\phi - D\delta\phi \\ &= (E - D\delta)\phi \end{aligned}$$

$$\int_{\delta_0}^{\delta} \frac{\delta}{(E - D\delta)} = \int_0^t \phi dt$$

$$\int_{\delta_0}^{\delta} \frac{A d\delta}{(E - D\delta)} = \int_0^t \phi dt$$

add

This can be further simplified:

Let,

$$u = E - D\delta \rightarrow du = -Dd\delta$$

$$\frac{du}{-D} = d\delta$$

and when,

$$\delta = \delta_0, u_0 = E - D\delta_0$$

$$\delta = \delta, u = E - D\delta$$

Therefore,

$$-\frac{1}{D} \int_{u_0}^u \frac{du}{u} = \int_0^t \phi \, dt$$

$$-\frac{1}{D} \int_{u_0}^u \frac{du}{u}$$

$$-\frac{1}{D} \ln u/u_0 = \phi t$$

$$\rightarrow \ln u/u_0 = -D\phi t$$

$$\ln \frac{u}{u_0} = -\frac{D}{A} \phi t$$

Substituting back in,

$$\ln \left(\frac{E-D\delta}{E-D\delta_0} \right) = -D\phi t$$

$$-\frac{D}{A}$$

$$\frac{E-D\delta}{E-D\delta_0} = \exp(-D\phi t) \leftarrow \exp\left(-\frac{D}{A} \phi t\right)$$

$$\delta = \frac{E}{D} - \frac{(E-D\delta_0)}{D} \exp(-D\phi t)$$

or, as equation (2.17) in the main text,

$$\delta = C_1 - C_2 \exp(-C_3 \phi t)$$

where,

$$C_1 = \frac{E}{D} = \frac{\frac{1}{3} \frac{A}{t} + K_g G_d}{K_c C_d (1-\nu) \frac{t}{\ell^2}}$$

$$C_2 = C_1 - \delta_0$$

$$C_3 = D = K_c C_d \frac{E}{(1-\nu)}$$

$$C_3 = \frac{D}{A}$$

where it is shown that the saturation in the deflection of the cantilever-beam specimen during neon irradiation is due to the texture dependent creep component.

Appendix E

Correlation of 100 KeV Neon Ion and Fission

Neutron Irradiation of Zirconium

In cascades with a high defect density, the Kinchen-Pease model tends to overestimate the number of Frenkel pairs which survive to alter the physical properties of the irradiated material since the recombination of displaced atoms with vacant lattice sites is neglected. This is a particularly important consideration in the simulation of neutron damage by 100 KeV neon ion irradiation as the defect densities of the neutron and ion damage are quite different (125). The ion damage cascade will extend along the trajectory of the neon ion whose range (~ 100 nm) (120) will be a measure of the longitudinal dimension of the cascade. For 100 KeV neon ions, the transverse dimension of the cascade is about 85% of its longitudinal dimension (121) leading to a low defect, cylindrical cascade. The situation is quite different in the case of fission neutron irradiation where the cascades are localized and are of high defect density. These differences are shown in Figure 1E and are tabulated in Table 1E. It is reasonable to expect more direct recombination of vacancies and displaced atoms during de-excitation of the neutron damage cascade as, from Table A1, it has a defect density of $\sim 10^3$ times that of the ion damage cascade.

Many attempts have been made to estimate the amount of this recombination which depends on the initial size, shape

and defect density of the cascade. Both Altovski (130) and Foreman (131) have independently performed computer calculations of the direct recombination of defects in a cascade using diffusion theory rather than a detailed atomic model (124). Altovskii concludes that 90% of the Frenkel pairs recombine in neutron irradiated Au for which $E_d = 25$ eV and the average PKO energy is 10 KeV. Foreman's calculations conclude that recombination is dependent on N_v^2 , where v is the initial vacancy density of the cascade and N is proportional to the initial spread of the vacancies. For the cascades described in Table A1, with N taken as the width of the cascade:

$$\frac{(N^2 v)_{\text{neutron}}}{(N^2 v)_{\text{ion}}} = \frac{(62)^2 \times 2.5 \times 10^{-2}}{(850)^2 \times 1.6 \times 10^{-5}} = 8.3 \quad (\text{E.1})$$

so that, from Foreman's model, recombination in the defect-rich fission neutron cascade is 8.3 times that in the 100 KeV neon ion cascade. The neutron and ion fluences are therefore required to generate the same number of displacements per atom, from Table A1, given by:

$$8.3 \times 136 \times 5.7 \times 10^{-28} \times 4.32 \times 10^{22} \phi_n = 401 \phi \text{ ion} \quad (\text{E.2})$$

from which,

$$\phi_n = 1.44 \times 10^4 \phi \text{ ion}$$

This correlation factor should be used in the analysis of the cantilever-beam results.

Table E1
Comparison of Neutron and Ion Damage Parameters in Zr (125)

	Damage Energy (KeV)	Average Number of Displaced Atoms per Cascade	Number of Cascades (cm ⁻³)	Cascade Dimensions		Defect Density Frenkel Pairs/Atom
				Longitudinal	Transverse	
100 KeV neon ion	40	401		100	85	1.6 x 10 ⁻⁵
fission neutron E>0.1MeV	--	136 ⁺	5.7x10 ⁻²⁸ N _O φ _n ⁺	6.2	6.2	2.5 x 10 ⁻²

N_O = 4.32 x 10²² Zr atoms cm⁻³

+ = Reference (132)

++ = Reference (120)

φ_n = neutron flux

PKO = primary knock-on

ε^{*}_d = 40 eV (32) the displacement threshold in Zr averaged over solid angle

Figure E1 Comparison of the size and shape of neutron and 100KeV neon ion irradiation.

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PUBLICATIONS

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