SUMMARY REPORT

Mechanical Properties of Neutron Irradiated Fuel Cladding Tubes

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There was few post irradiation examination data on the mechanical properties of domestic fuel cladding tubes used for light water reactors, then those data obtained abroad have been often used in the fuel design or fuel performance codes. Although, many reports discussed the deformation mechanism of the tube, almost all the data were not obtained from irradiated specimens but unirradiated ones. In recent years, systematic post irradiation examinations on domestic fuel elements used in Japanese light water reactors and the related studies were performed.

This report first summarizes briefly the crystallographic texture which characterizes the properties of Zircaloy fuel cladding tubes, followed by an explanation of basic properties such as elasticity, plasticity, creep and fatigue. Finally, the up-to-date results are introduced.

KEYWORDS: review, fuel cladding, Zircaloy, neutron beams, irradiation, mechanical properties, elasticity, plasticity, creep, fatigue

I. INTRODUCTION

The strength and deformation properties of fuel cladding tubes not only provide a basis for fuel design, but are also important as input data of fuel performance code. As to the mechanical properties of irradiated Zircaloy fuel cladding tubes used for commercial light water reactors, there are only limited post irradiation test data on domestic claddings, then those obtained abroad have been often used instead. In particular, strength and plastic deformation depend greatly on crystallographic texture i.e. preferred orientation, and tend to be affected by production processes, so that the acquisition of data on domestic claddings has long been awaited. Although there are many reports describing the deformation properties of cladding tubes, most of them evaluate unirradiated specimens, with few report using specimens prepared from irradiated cladding tubes.

This report first describes briefly the crystallographic texture that characterizes Zircaloy fuel cladding tubes, followed by an explanation of basic properties such as elasticity, plasticity, creep, fatigue etc. Finally, up-to-date results of studies are introduced.

II. DEFORMATION SYSTEMS AND CRYSTALLOGRAPHIC TEXTURE

In the tension and compression tests for zirconium single crystals {1012}, {1121}, {1122} and {1123}, twins have been observed(1). On the other hand, slip mostly occurs along {1010}; in fact when the shearing stress of a basal plane was nine times(2) or about 25 times(3) as great as that of a prismatic plane, slip still occurred along {1010}; in fact when the shearing stress of a basal plane was nine times(3) or about 25 times(3) as great as that of a prismatic plane, slip still occurred along {1010}. Figure 1(a)~(c)(3) summarizes the information obtained data on the deformation systems.

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Oarai-machi, Ibaraki-ken 311-13.
of α-Zr and its alloys.

Since the number of deformation systems is small and their distribution is asymmetrical as described above, if α-Zr and its alloys are subjected to deformation working, then the basal planes are oriented in parallel with the deformation direction, resulting in the formation of strong deformation textures. This orientation model changes slightly in accordance with c/a ratios (4)~(9). The deformation textures of zirconium and Zircaloy are formed by complicated interactions between slip and twins (10)(11). The factor determining the textures of fuel cladding tubes is the relative ratio \((Q = Rw/R_D)\) of the wall thickness reduction rate \((Rw)\) to the diameter reduction rate \((R_D)\) (12)(13). The relationships between \(Rw/R_D\) (Q value) and textures generated in fuel clad- dings can be summarized as shown in Fig. 2 (14). These indicate, for example, that in the case of working conditions in which the wall thickness reduction rate is greater than the diameter reduction rate \((Rw/R_D > 1)\), strain in the longitudinal direction of a tube is greater than that in the tangential direction with C axes centralizing from the radial to the tangential direction by ±30 to 50°.

![Fig. 1 Crystallographic planes and deformation systems of α-Zr](image)

**Fig. 1** Crystallographic planes and deformation systems of α-Zr (3) (a) Slip systems to \(<1\overline{2}10>\) direction, (b) Slip systems to \(<2\overline{1}1\overline{3}>\), (c) Twinning systems

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<table>
<thead>
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<th>TYPE OF DEFORMATION</th>
<th>TUBE REDUCTION WITH</th>
<th>TUBE</th>
<th>STRAIN ELLIPSE IN THE PLANE TO THE DIRECTION OF DEFORMATION</th>
<th>DEFORMATION TEXTURE</th>
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<td>RW/R_D &gt; 1</td>
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<td>(0002) POLE FIGURE</td>
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**Fig. 2** Strain distribution and resulting textures of tubes (14)
III. MECHANICAL PROPERTIES

1. Elastic Properties

When the power of the fuel elements is raised, the gaps between claddings and pellets are closed, and the tubes are subjected to a force as if they were pushed from inside. To predict the stress generated in the claddings, elastic moduli are required as basic data. In stress analysis, isotropic elastic moduli are generally assumed. In the case of claddings, however, anisotropic elastic moduli due to textures must be taken into account, and there are several reports\(^{(15)-(20)}\) describing the relationships between crystallographic textures and elastic moduli.

The models developed by Voigt\(^{(21)}\) and Reuss\(^{(22)}\) are well known as techniques for obtaining the mean elastic modulus of a polycrystal from that of a single crystal. While the elastic deformation of polycrystals has been reported as close to the Reuss model\(^{(23)}\), many experimental values are in fact between the two models\(^{(24)}\). As to the methods of obtaining relationships between the elastic modulus of a polycrystal with textures and the texture parameters, some papers\(^{(25),(26)}\) represent a polycrystal with single crystal lying in the orientation to which the C axes are most centralized. In more generalized single crystal methods, the elastic moduli\(^{(27)}\) of the single crystals in various orientations of a polycrystal are weighted with the relative volume of the single crystals in each orientation and integrated\(^{(28)}\) with regard to all the orientations. Additionally, there is a simple method\(^{(29)}\) enabling the elastic anisotropy of fuel claddings to be calculated by using \(f\) values\(^{(30)}\).

According to the results (Figs. 3 and 4) calculated in detail on the elastic anisotropy of typical BWR fuel claddings, the elastic modulus is nearly isotropic at room temperature, but anisotropy increases monotonously as temperature rises, and the effects of changes in texture on elastic moduli determined by the longitudinal uniaxial tension of claddings hardly appear\(^{(39)}\).

There are two experimental methods; one is the static measurement method in which the relation between stress and strain is first obtained by using a strain gauge, followed by measurement of the gradient of the stress-strain diagram, and the other is the dynamic measurement method in which the elastic moduli is obtained from the resonance\(^{(31)}\) of a rod specimen and acoustic velocity\(^{(32)}\). Measured elastic moduli of various Zr alloys (Zircaloy-2 (Zry-2), Zr-Cr-Fe and Zr-2.5Nb) indicate that the effects of the difference in alloying elements were smaller than those of the difference in crystallographic textures, and that the effects of the difference in texture were greater than those of the difference (1,500 wt. ppm or less) in oxygen concentrations\(^{(33)}\). Moreover, although some papers\(^{(34),(35)}\) reported that the elastic moduli of irradiated Zircaloy claddings were several percent greater than those of unirradiated ones, additional tests are required for confirmation.

2. Plastic Properties

1. Uniaxial Tensile Properties

Many longitudinal tension tests\(^{(36)-(39)}\) have been performed for fuel claddings to examine the mechanical properties of irradiated fuel claddings. Since strength and ductility depend not only on irradiation conditions but also on the crystallographic textures, shapes

\[
\begin{bmatrix}
\sigma_x \\
\sigma_y \\
\sigma_z \\
\tau_{xy} \\
\tau_{xz} \\
\tau_{yz}
\end{bmatrix}
= \begin{bmatrix}
\bar{S}_{11} & \bar{S}_{12} & \bar{S}_{13} & 0 & 0 & 0 \\
\bar{S}_{12} & \bar{S}_{22} & \bar{S}_{23} & 0 & 0 & 0 \\
\bar{S}_{13} & \bar{S}_{23} & \bar{S}_{33} & 0 & 0 & 0 \\
0 & 0 & 0 & \bar{S}_{44} & 0 & 0 \\
0 & 0 & 0 & 0 & \bar{S}_{55} & 0 \\
0 & 0 & 0 & 0 & 0 & \bar{S}_{66}
\end{bmatrix}
\begin{bmatrix}
\varepsilon_x \\
\varepsilon_y \\
\varepsilon_z \\
\gamma_{xy} \\
\gamma_{xz} \\
\gamma_{yz}
\end{bmatrix}
\]

Fig. 3 Definition of stress and strain in cladding tube\(^{(29)}\)
and dimensions of the specimens, the dispersion of measured values given in the past is relatively large.

The effects of neutron irradiation on the relation between stress and strain are remarkable. As shown in Fig. 5, the strain hardening exponents of the unirradiated materials are nearly constant, but those of the irradiated materials rapidly decrease as strain increases. Many studies were performed on this phenomenon because of its direct relation to the ductility of materials, and observation of slip lines and TEM observations indicated that the phenomenon was closely related to the localized deformation in slip band, with a further detailed study now in progress. The elongations of irradiated domestic Zry-2 tubes have been measured precisely and the proportional limits are shown in Fig. 6. The proportional limit means the stress from which dislocation movement starts, and it is a noteworthy value, especially for irradiated materials, because of its potential relationship to failure conditions of fuel tubes such as threshold stress for SCC or fatigue limit.

The effects of neutron irradiation on the increase in strength are evident. After 1-cycle irradiation in a reactor, 0.2% yield strength of Zry-4 (stress-relief-annealed) and of Zry-2 (recrystallization-annealed) increase by 10-20% and about 100-200%, respectively. The increase in yield strength is expressed as an exponential power of the neutron fluence.
and this exponent decreases from 1/2 to 1/3 as the amount of neutron fluence increased, and further decreased down to about 0.1 in Zry-2 (recrystallization-annealed) when irradiation exceeds $10^{24}$ n/m² ($E>1$ MeV). As shown in Fig. 7(a) and (b), the strength of irradiated fuel claddings hardly depends on final heat treatment conditions, but saturates for the amount of irradiation.

The temperature dependence of mechanical strength of the unirradiated materials, as shown in Fig. 8, decreases as temperature increases with a following further slower decrease, whereas in the irradiated materials the strength decreases as the testing temperature rises, but the decreasing rate tends to become smaller temporarily at about 300°C. This stems from the effect of post-irradiation annealing hardening; namely, at about 300°C, the diffusion of oxygen etc. becomes easy, and these interstitial impurities may be trapped by irradiation defects, thereby interfering with dislocation movement.

When strain rate increases from $5 \times 10^{-4}$ to $1 \times 10^{-1}$ min$^{-1}$, the strengths of both unirradiated and irradiated materials tend to rise. The strain rate sensitivity parameter $m$ for 0.2% yield strength for the unirradiated material was 0.037, and decreased to 0.021 for the irradiated material. In the case of the fuel claddings, as observed for many other alloys, a decrease in $m$ value reduces failure strain.

(2) Multi-axial Stress Properties

Bi-axial or multi-axial stress states occur in in-service fuel cladding tubes, and it is important to understand the strength characteristics of the cladding tubes under these stress conditions. Accordingly, and series of internal pressure burst tests have been carried out for unirradiated tubes and for tubes used as fuel claddings, and part of the results are shown in Fig. 9.

Because the yield conditions for fuel claddings under multi-axial stress tend to be affected by crystallographic textures, yield conditions have been obtained to date by tension/internal pressure tests, tension/torsion tests, and by Knoop hardness method. It is pointed out, however, that the yield conditions obtained from Knoop hardness sometimes do not coincide with those by the internal pressure method. Figure 10 shows example yield loci of typical unirradiated Zircaloy claddings under bi-axial stress state. Unlike the von Mises' yield condition, texture hardening is observed. Although
Texture hardening can be explained by using Hill's theory of anisotropy\textsuperscript{(58)-(60)}, yield locus depends on temperatures and the amount of deformation\textsuperscript{(60)}, so all the conditions cannot always be represented by Hill's conventional theory. Naturally, the yield loci depend on the crystallographic texture of a fuel cladding, and anisotropy appears more strongly on the yield locus as the C axis is concentrated to the radial direction of the fuel cladding\textsuperscript{(60)}.

From the results of experiments carried out for unirradiated materials, it is widely known that the yield conditions of fuel claddings are accompanied by strong anisotropy. Figure 11\textsuperscript{(48)} shows the yield conditions under bi-axial stress by combining the results of tension tests performed for irradiated domestic fuel claddings with those of internal pressure burst tests. Available data on the unirradiated materials have strong anisotropy, and are deviated from the von Mises's curve representing the yield conditions of isotropic materials. However, data on the irradiated materials, according to recent work\textsuperscript{(48)}, nearly coincide with von Mises's yield condition.

![Graph](image-url)
revealing that the yield condition of fuel claddings approach those of isotropic materials once the claddings are irradiated.

From the irradiation dependence of Knoop hardness, used to examine in detail the deformation behavior of irradiated fuel claddings under multi-axial stress, it was found that the anisotropy of yield conditions, even under any bi-axial stress state except that in internal pressure burst tests, was mitigated, and that this phenomenon stemmed from the fact that irradiation defects caused by neutron irradiation showed strong resistance against the slip along the prismatic planes of \( \alpha \)-Zr in comparison with other deformation systems\(^{(67)}\).

As to the analytical study on the effects of crystallographic textures and deformation systems on the deformation behavior of fuel
claddings under multi-axial stress states, a few papers\(^{(56)-(57)}\) have represented the macroscopic deformation of fuel claddings by the slip and twin deformation of \(\alpha\)-Zr single crystals in the maximum abundance direction. In this concept, the macroscopic stress generated in the fuel cladding is converted into stress acting on the single crystals inclining toward a given direction in the cladding, and deformation is considered to take place when the resolved shear stress exceeds the critical resolved shear stress for slip or twin. As examples, the paper\(^{(48)}\) using Tailor’s theory for the plastic deformation of polycrystals and those\(^{(49)-(50)}\) using other approaches are available.

Based on the studies explained above, the numerical model\(^{(71)}\) which can predict the yield conditions of fuel claddings taking into account the crystallographic textures and critical resolved shear stress of \(\alpha\)-Zr was reported, followed by the explanation for mitigating the anisotropy of yield locus observed in the tests for unirradiated materials as shown in Fig. 12.

Many studies\(^{(58)-(44)-(66)-(73)-(76)}\) have been carried out on the ductility of fuel claddings. For isotropic materials, the role of stress states for ductility was examined in detail\(^{(77)}\), and it was seen that ductility becomes lower in multi-axial stress states. In Zr-alloy tubes, the minimum uniform elongation is shown at \(\alpha\) (longitudinal stress/circumferential stress) = 1/2\(^{(47)}\) or \(\alpha = 2/3\(^{(74)}\). A numerical analysis which was able to predict the ductility from the stress states of anisotropic materials was also reported\(^{(58)}\). The elongation of the un-irradiated materials (recrystallization-annealed) is as large as 50% or more. Although this value decreases with the increasing amount of neutron irradiation, it becomes constant at irradiations as high as \(5 \times 10^{25} \text{ n/m}^2\), and is more than 1%.

3. Creep Properties

The features of in-reactor creep deformation are shown in Fig. 13. This includes the deformation due to neutron flux as well as thermal creep. The relation between neutron flux \(\phi \ (E > 1 \text{ MeV})\) and creep strain rate \(\dot{\varepsilon}\) is
often represented by $\dot{\epsilon} = \dot{\phi}^P$, with some reports assuming $P=0.7^{(78)}$, $0.85^{(78)}$ and $1.0^{(78)}$. In many uni-axial tests ($573 \text{ K}, 0.6\sim2.5 \times 10^{17} \text{n/m}^2 \cdot \text{s}$), $P=1.0 \pm 0.1$, whereas from the data for cold-worked Zry-2 ($573 \text{ K}$), $P=0.5^{(80)}$. In the formulation based on these data, $P=1$ has been frequently employed.

The stress dependence of in-reactor creep has been reported$^{(81)}$~$^{(86)}$ in a model explaining the irradiation creep mechanism. An example is given in Fig. 14. The amount of creep strain in the low stress region in which irradiation growth is dominant is negligibly small; accordingly, the strain rate in the intermediate stress region is important from the viewpoint of fuel behavior analysis. In this region, creep deformation advances through two steps; one is the step in which dislocation climbs to overcome the obstacle taking place due to irradiation defects or work hardening, and the other is the subsequent slip step. In the lowest stress zone within the intermediate stress region, it is predicted that the dislocation climb motion is accelerated by irradiation.
and there may be a region where creep rate increases stepwisely as if there were no obstacle caused by irradiation\(^{(86)}\). On the other hand, many data indicate that stress dependence is not stepwise, but increases continuously with the increase of stress\(^{(87)}\)(\(^{(88)}\). When the temperature becomes higher, dislocation tends to slip, and the climb rate of dislocation becomes relatively slower, so the rate-determining step is changed and the step in which obstacle is overcome by the climb of dislocation dominates creep deformation.

The ductility at the in-reactor creep rupture is also affected by neutron irradiation. As mentioned previously, the out-of-pile tension tests of irradiated materials showed a remarkable decrease in failure elongation, whereas the failure elongation for in-reactor creep was sometimes greater than that for out-of-reactor creep of unirradiated materials\(^{(89)}\). The strain rate sensitivity parameter \((m\) value in Fig. 14\) under irradiation tends to become greater than that of out-of-reactor. Since the failure elongation of a material generally increases with the increasing \(m\) value as shown in Fig. 15, the strain which starts accelerating creep in reactors is greater than that of out-of-reactor tests for unirradiated materials\(^{(89)}\).

\[\text{Fig. 15 Relationship between failure elongation and strain rate sensitivity parameter for various materials}^{(90)}\]

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure15.png}
\caption{Relationship between failure elongation and strain rate sensitivity parameter for various materials\(^{(90)}\)}
\end{figure}

4. Fatigue Properties

Zircaloy claddings are subjected to repeated loadings as the power of fuels changes during operation. The fatigue strength of zirconium alloys under repeated loadings have been studied from various viewpoints. As to unirradiated materials, the fatigue damage \(^{(90)}\)(\(^{(91)}\) of \(\alpha\)-Zr and Zry-2, or the high-cycle fatigue characteristics\(^{(92)}\)(\(^{(93)}\) of Zr-2.5Nb alloy were reported. In addition, many studies were reported on fatigue strength of Zry-2; they included Zry-2\(^{(94)}\) containing relatively large amount of hydride, the weids of Zry-2\(^{(96)}\) and Zry-2 rods\(^{(97)}\) containing several level of oxygen concentration ranging 140~1,740 ppm. Investigation was carried out on the effects of temperatures, atmospheres (air/vacuum) and cold working on the fatigue lives of crystal bar Zr and Zry-2, with the fatigue strength \((10^6\) cycles) at 300°C being got as 109 and 197 MN/m\(^2\)\(^{(97)}\).

With regard to the effects of neutron irradiation on the fatigue properties of irradiated Zry-2, it was reported\(^{(98)}\), on the basis of the bending tests for Zry-2 sheets irradiated up to \(1.3\sim2.6\times10^{24}\) n/m\(^2\) in reactor water, that in the range of \(\varepsilon_i>1\%\) and \(N_f<10^6\) fatigue life did not change whether sheets were irradiated or not. In the study in which crack-propagation rates were measured with CT specimens (Zry-2) irradiated in a reactor, no effect due to irradiation on crack-propagation rate was observed\(^{(99)}\). Also, it was reported\(^{(100)}\), in accordance with the tests on Zry-2 subjected to \(5.5\times10^{25}\) n/m\(^2\) fast neutron irradiation, that the failure cycles of Zry-2 was shortened by about 1/2 on the low cycle side \((N_f<10^4)\), but prolonged a little on the high cycle side \((N_f<10^6)\).

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure16.png}
\caption{Results of fatigue tests on irradiated fuel claddings. With the total strain amplitude \(\varepsilon_t\) of about 0.3\% \((N_f<10^5)\) being the boundary, the life of the irradiated material was longer than that of the unirradiated on the high strain amplitude side \((\varepsilon_t>0.3\%)\), whereas the former was shorter than the latter on the low strain amplitude side \((\varepsilon_t<0.3\%)\), and the fatigue limit was about 0.18\%. The effects of irradiation of fatigue}

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life can be interpreted as follows: In a high strain amplitude, crack propagation rate $C$ depends on the width of the plastic region at the end of the crack. As the strength of a material increases, the plastic region narrows, and the $C$ value decreases, for example, with $C \propto \sigma_y^{-3}$ ($^{193}$). Since $\sigma_y$ increases with the increase of the amount of irradiation $^{135}$, crack propagation rates of an irradiated material are lower than those of unirradiated materials at high strain amplitude. On the other hand, fatigue damage accumulates during irreversible slip at a low strain amplitude, so the strain corresponding to the proportional limit may get close to the fatigue limit. The strain corresponding to the proportional limit $^{195}$ of irradiated Zry-2 is also shown in Fig. 16. This value nearly coincides with the fatigue limit of the irradiated Zry-2 cladding. Fatigue damage, moreover, is sensitive to the defect properties of materials such as stacking fault energy or micro structures, and it is well known that the effects of irradiation on fatigue life are also unstable for other structural materials $^{193}$, $^{194}$.

**IV. CONCLUSIONS**

This paper reviews the macroscopic mechanical properties of neutron irradiated fuel claddings. To understand these phenomena more clearly, research relating macroscopic phenomena to microscopic behavior as well as research on irradiation defect characteristics are necessary. In addition, preparation of a database is essential to utilize the information acquired to date.

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