Thermal Creep of Zircaloy-4 Cladding under Internal Pressure

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Creep characteristics of Zircaloy-4 stress-releaved cladding under internal pressure were studied. Creep tests were conducted under 21 conditions chosen from the temperature range of 603~693 K and the hoop stress of 49~314 MPa. The maximum accumulated test period was 3,000 h. Diametral creep data were analyzed by separating the primary (transient) and the secondary (steady-state) creep, based on Dorn's quasitheretical model, and the following equations were derived:

Total creep strain: \[ \varepsilon = \varepsilon_p [1 - \exp(-52(\varepsilon/p)^{4/3})] + \varepsilon_s, \]

Saturated primary creep strain: \[ \varepsilon_p = 2.16 \times 10^{-3}(\varepsilon_s)^{0.16}, \]

Steady-state creep rate: \[ \dot{\varepsilon}_s = 5.7 \times 10^{14}(E/T) [\sinh(1.13 \times 10^9 \sigma/E)]^{3/2} \exp(-2.72 \times 10^9 /RT). \]

The apparent activation energy of the steady-state creep, which is 2.72 \times 10^5 J/mol, is in good agreement with those of self-diffusion of Zr in Zr-Sn alloys and suggests the self-diffusion is the control mechanism.

KEYWORDS: activation energy, cladding, internal pressure, pressure dependence, self-diffusion, stress analysis, stresses, thermal creep, Zircaloy-4

I. INTRODUCTION

Zircaloy cladding is widely used in thermal reactors under various stress and thermal conditions. In PWR's, the cladding is under the external pressure by coolant so that diameter creeps down to become contact with the fuel pellets. Thermal expansion of pellets in power ramp or pellet swelling generates internal pressure in the cladding so as to increase diameter. Under PCMI (Pellet-Clad Mechanical Interaction) conditions, the cladding may be stressed in the axial direction. So the creep property is one of the essential characteristics of the cladding in predicting the fuel rod performance, and many studies have been reported(1)~(12). But in most cases, the materials studied are pressure tubes for heavy water reactors, which means the material conditions, heat treatment, manufacturing process, texture etc. are different from those of current cladding tubes. Also the applicability of the proposed creep equations are limited to specific conditions as to temperature and stress range. There is few systematic research on the creep properties of cladding tubes in the wide range of condition. A series of studies by Murty et al.(1)~(3) are, however, systematic and they have proposed not only creep equations under constant conditions but also equations considering recovery, anisotropy etc. But these equations do not seem to be applicable in some conditions, especially in low stress region.

In order to comprehend the complete creep characteristics of Zircaloy cladding tubes, their creep behavior under various conditions, e.g. internal pressure, external pressure and/or axial stress are to be known. Creep under external pressure is, however, known to be very complicated because it is affected by dimensions of the tube specimen (ovality, wall thickness variation, axial support length etc.)(13)(14). As to the axial creep property, large anisotropy against the biaxial creep is reported(15).

Then, in this study, biaxial creep tests under internal pressure were conducted to establish the fundamental creep characteristics of Zircaloy cladding tubes applicable for wide range of conditions.

II. MATERIAL

Specimens for the creep tests were cut out from a lot of cold-worked and stress-relieved...
Zircaloy-4 cladding tubes which had been manufactured in the usual mass-production process. The nominal dimensions are 10.72 mm O.D. x 9.48 mm I.D. x 0.62 mm t. The ingot chemistry is shown in Table 1, and the basal properties in Table 2.

### Table 1 Ingot chemistry

<table>
<thead>
<tr>
<th>Element</th>
<th>Sn</th>
<th>Fe</th>
<th>Cr</th>
<th>Fe+Cr</th>
<th>O</th>
<th>C</th>
<th>Zr</th>
</tr>
</thead>
<tbody>
<tr>
<td>Composition (wt%)</td>
<td>1.43~1.52</td>
<td>0.20~0.21</td>
<td>0.10~0.11</td>
<td>0.30~0.32</td>
<td>0.130</td>
<td>0.008~0.015</td>
<td>Bal.</td>
</tr>
</tbody>
</table>

### Table 2 Basal properties of material tested

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>UTS (MPa) at 658 K</td>
<td>453 456</td>
</tr>
<tr>
<td>0.2% Y.S. (MPa)</td>
<td>397 395</td>
</tr>
<tr>
<td>Elongation (%)</td>
<td>22.2 21.3</td>
</tr>
<tr>
<td>Hydrogen (ppm)</td>
<td>≤0.20</td>
</tr>
<tr>
<td>Nitrogen (ppm)</td>
<td>14 16</td>
</tr>
<tr>
<td>Oxygen (ppm)</td>
<td>48 56</td>
</tr>
<tr>
<td>Nitrogen (ppm)</td>
<td>1,280 1,350</td>
</tr>
</tbody>
</table>

## III. EXPERIMENTAL

### 1. Test Apparatus and Specimen

Figure 1 shows the schematic diagram of the test apparatus. It consists of six high-pressure supply systems, three electric furnaces, temperature control systems and recording systems. Specimens are connected to the high-pressure systems, then set in the electric furnace and kept at constant temperature under internal inert gas pressure and the inert gas atmosphere. For measuring the creep strain, specimens are cooled down and took off from the test apparatus. Each internal pressure supply system has an accumulator kept at a constant temperature, which enables the test pressure kept within ±0.5%. Electric furnaces can be moved down quickly at the end of the test period, which can make the specimen temperature down fast to avoid the recovery of the creep strain.

Zircaloy plugs are welded at both ends of the specimen, one of which can be connected to the high-pressure system with a Swagelok type fitting. A stainless steel mandrel is used to minimize the void volume and to avoid bowing of the specimen during the test.

### 2. Test Procedure

After the specimens are connected to the
high-pressure system, they are pressurized by internal pressure to just below the test pressure with Ar gas at room temperature. Then the specimens are heated up to the test temperature, by moving up the pre-heated furnace. Test pressure is adjusted after the test temperature has been attained. Specimens are cooled down periodically and disconnected from the high-pressure system to measure the diametric creep strain and the axial elongation with a micrometer or a comparator, respectively. The accuracy of the measurements are estimated to be ±2 μm.

The axial strain was measured using four 50 mm gauges scribed at 90° interval.

3. Test Conditions

The nominal test conditions adopted in this study are summarized in Table 3. Nineteen (19) combinations of temperature and hoop stress were chosen from the temperature range of 603 ~ 693 K and the hoop stress of 78 ~ 314 MPa. Two (2) additional low stress tests were conducted at 693 K, which would be described in detail in 5.1. Figures in Table 3 indicate the accumulated creep test time for each condition. The hoop stress is calculated using the measured dimensions of the specimen and the average internal pressure during the test as

$$\sigma_0 = \frac{PD}{2t},$$

where $$\sigma_0$$: Hoop stress

$$P$$: Internal pressure

$$D$$: Mean diameter

$$t$$: Average thickness.

<table>
<thead>
<tr>
<th>Hoop stress (MPa)</th>
<th>78</th>
<th>118</th>
<th>157</th>
<th>196</th>
<th>235</th>
<th>275</th>
<th>314</th>
</tr>
</thead>
<tbody>
<tr>
<td>Internal pressure (MPa)</td>
<td>9.6</td>
<td>14.5</td>
<td>19.3</td>
<td>24.1</td>
<td>28.9</td>
<td>33.8</td>
<td>38.6</td>
</tr>
<tr>
<td>Temperature (K)</td>
<td>603</td>
<td>633</td>
<td>663</td>
<td>693</td>
<td>3,000</td>
<td>3,000</td>
<td>3,000</td>
</tr>
<tr>
<td>603</td>
<td>3,000</td>
<td>1,920</td>
<td>960</td>
<td>480</td>
<td>480</td>
<td></td>
<td></td>
</tr>
<tr>
<td>633</td>
<td>960</td>
<td>960</td>
<td>240</td>
<td>240</td>
<td>240</td>
<td></td>
<td></td>
</tr>
<tr>
<td>663</td>
<td>480</td>
<td>480</td>
<td>120</td>
<td>120</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

IV. RESULT

Creep strains as function of time at 633 K are shown in Fig. 2 as an example. In the log-log plot, time vs. strain curves seem to be straight but the slope increases as the stress goes higher.

Figure 3 shows the variation of the circumferential profile with time. It is obvious that the creep deformation proceeds uniformly up to more than 10% of strain.

The axial strain was so small in comparison with the diametric one (1/100 to 1/20) as to be regarded as zero.

V. ANALYSIS AND DISCUSSION

Obquvist et al. (15) analyzed the creep deformation of isotropic thin-walled cylindrical material under the internal pressure and showed that it did not deform in axial direction. The results of this study are consistent with it, although some literature reported the anisotropy of creep deformation of Zircaloy (16)~(18).

Various types of equations have been proposed for the creep deformation of Zircaloy (1)~(3) (9)~(12). Figure 2 shows the linear relationship between the creep strain ε and the time t on the log-log plot, which means that ε can be formu-
lated as a function of time as follows:
\[ \varepsilon = k(T, \sigma) t^m, \quad m = f(T, \sigma), \]

\( K \) and \( m \) are presumed to be rather complex functions of stress \( \sigma \) and temperature \( T \), but this type of formula has no clear physical meaning, nor correlation with creep mechanism. Then, in this study, creep data were analyzed by separating the primary (transient) and the secondary (steady-state) creep. First, the steady-state creep rate was decided from the data which seemed to have attained the steady-state using the least square method and it was extrapolated to \( t=0 \) to calculate the saturated primary creep strain. Table 4 summarizes the results together with the average test temperatures and the applied hoop stresses.

The model creep equation employed for the analysis of the results in this study is Dorn's quasitheoretical one(19)(20), which is also the basic model of Murty et al.'s creep equation(1):

\[ D = D_0 \exp(-Q/RT); \]

\[ E: \text{Elastic modulous (MPa)} \]

\[ \text{Diffusion coefficient (m}^2/\text{h)} \]

\[ Q: \text{Activation energy (J/mol)} \]

\[ R: \text{Gas constant (=8.3169 J/mol-K)} \]

\[ b: \text{Burger's vector (m)} \]

\[ k: \text{Boltzmann constant (=1.380x10^{-23} J/K)} \]

\[ A,B,K: \text{Constants}. \]

1. Steady-state Creep

From Eq. (3), steady-state creep rate can be formulated as

\[ \dot{\varepsilon}_s = A' \frac{E}{T} \exp\left(\frac{B}{E} \sigma_0/E\right) \exp\left(-\frac{Q}{RT}\right), \quad (4) \]

where \( A' = A \left( E b/k T \right) D_0 \).

This equation shows that, at a constant temperature, \( \dot{\varepsilon}_s \) and \( \sigma_0/E \) shall be on a straight line in the semi-logarithmic plot (see Fig. 4), where \( E \) is calculated as follows(3):

\[ E = 1.148 \times 10^9 - 5.99 \times 10^8 T \text{ (MPa)} \]

\[ B \text{ in Eq. (4) was determined from the slopes in Fig. 4 as } B=2.40 \times 10^3. \]

Constant \( C \) and the apparent activation energy \( Q \) were determined by \( 1/T \) vs. \( (\ln \dot{\varepsilon}_s - \ln E/T - 2.40 \times 10^3 \sigma_0/E) \) plot (see Fig. 5):

\[ A' = 3.62 \times 10^{11}, \quad Q = 2.72 \times 10^6 \text{ (J/mol)}. \]

Then, Eq. (4) becomes

\[ \dot{\varepsilon}_s = 3.62 \times 10^{11} \left( E/T \right) \exp(2.40 \times 10^3 \sigma_0/E) \cdot \exp(-2.72 \times 10^6/RT). \quad (4') \]
The apparent activation energy $Q = 2.72 \times 10^5$ J/mol is in good agreement with those of the self-diffusion of Zr-Sn alloys reported by Lyashenko(21), which were $2.60 \times 10^5$ J/mol (62.0 kcal/mol) for Zr-1.30%Sn and $3.14 \times 10^5$ J/mol (75.0 kcal/mol) for Zr-2.39%Sn. This result suggests that the self-diffusion is the rate controlling step in the steady-state creep of Zircaloy.

It is evident, however, that Eq. (4) is not applicable for low stress levels, for the stress dependency of the steady-state creep rate is expressed by an exponential term, which has positive value when the stress approaches to zero. Garofalo(22) showed the stress dependency of the steady-state creep rate was different in high stress region and low stress region. In high stress region, it is expressed as

$$\dot{\varepsilon}_s = C \exp(\beta \sigma). \quad (6)$$

On the other hand, in low stress region

$$\dot{\varepsilon}_s = C' \sigma^n. \quad (7)$$

These two equations can be combined as

$$\dot{\varepsilon}_s = C''(\sinh \alpha \sigma)^n, \quad (8)$$

where $\alpha = \beta / n$, $C' = C'' \alpha^n$, $C'' = C2^n$.

Here, “low stress” means $\sigma < 0.8$ and “high stress” $\sigma > 1.2$. Stress levels adopted in this study (see Table 3) are in the “high stress” range. In order to study the stress dependency of $\dot{\varepsilon}_s$ in low stress region, two sets of additional creep tests were conducted. First, specimens were creep strained about 1.4% under “high” stress: 120 h at 693 K, 118 MPa, to make the primary creep strain saturated, then the stress were lowered to 49 or 59 MPa. After holding these conditions for 48 h to avoid the effect of strain recovery, creep strains were measured every 240 h and steady-state creep rates were determined. The results are shown in Table 5. Log-log plot of these data vs. $\sigma/E$ shows the stress exponent

<table>
<thead>
<tr>
<th>Temperature (K)</th>
<th>Hoop stress (MPa)</th>
<th>Steady-state creep rate (h⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td>693.5</td>
<td>48.8</td>
<td>$3.68 \times 10^{-6}$</td>
</tr>
<tr>
<td>693.4</td>
<td>57.7</td>
<td>$5.42 \times 10^{-6}$</td>
</tr>
</tbody>
</table>
in Eq. (7) is 2.1. Therefore, Eq. (8) becomes
\[ \dot{\varepsilon} = 1.57 \times 10^{13} (E/T) \sinh(1.13 \times 10^5 \sigma/E) \exp(-2.72 \times 10^{3}/RT). \] (9)

Murty et al. (23) reported the value of 5.3 for the stress exponent in case of axial creep of Zircaloy, which was much larger compared to this study.

Figure 6 shows the comparison of the predicted steady-state creep rate by Eq. (9) and the experimental results, which are in good agreement including low stress data.

2. Transient Creep

The relationship between the saturated primary creep strain \( \varepsilon_p \) (shown in Table 4) and the steady-state creep rate \( \dot{\varepsilon}_{\text{sat}} \) predicted by Eq. (4) is shown in Fig. 7, which can be formulated using the least square method as
\[ \varepsilon_p = 2.16 \times 10^{-2} (\dot{\varepsilon}_{\text{sat}})^{0.109}. \] (10)

In the analysis of the time dependency of the primary creep, the primary creep strain \( \varepsilon_p \) was estimated by subtracting the steady-state creep strain from the total strain \( \varepsilon \);
\[ \varepsilon_p = \varepsilon - \dot{\varepsilon}_{\text{sat}} t. \]

Assuming that Eq. (4) is applicable for the creep of Zircaloy,
\[ \varepsilon_p = \varepsilon_p \{1 - \exp(-K \dot{\varepsilon}_{\text{sat}} t)\}, \] (11)
\[ \varepsilon_p / \varepsilon_p = 1 - \exp(-K \dot{\varepsilon}_{\text{sat}} t). \] (11')

Then, \( \dot{\varepsilon}_{\text{sat}} t \) shall have the linear relationship between \( \ln(1 - \varepsilon_p / \varepsilon_p) \) in a constant (temperature and stress) condition. It is not true (see Fig. 8), but \( (\dot{\varepsilon}_{\text{sat}} t)^{0.5} \) vs. \( \ln(1 - \varepsilon_p / \varepsilon_p) \) is found to be linear (see Fig. 9). Then,
\[ \varepsilon_p = \varepsilon_p \{1 - \exp(-K (\dot{\varepsilon}_{\text{sat}} t)^{0.5})\}, \] (12)

\( K \) is a complex function of the test conditions (temperature and stress), but it will not be a cause of the large error if a constant value is used for \( K \) to avoid the complexity of the equation. \( K \) is estimated to be 52. Then finally, the total creep strain is formulated as follows;

![Fig. 6 Comparison of predicted steady-state creep rates with measured values](image)

![Fig. 7 Relationship between saturated primary creep strain and steady-state creep rate](image)
Figure 8 Relationship between $\dot{\varepsilon}_t$ and $\ln(1-\varepsilon_p/\dot{\varepsilon})$.

\[
\frac{\dot{\varepsilon}_t}{t} \times 10^{11}
\]

\*\*\*\*

Figure 9 Relationship between $\sqrt{\dot{\varepsilon}_t}$ and $\ln(1-\varepsilon_p/\dot{\varepsilon})$.

\[
\frac{\sqrt{\dot{\varepsilon}_t}}{t} \times 10^{11}
\]

\*\*\*\*

\[\dot{\varepsilon}_t = \frac{\dot{\varepsilon}}{52(\dot{\varepsilon}_{\text{crit}})^{0.5}} + \dot{\varepsilon}_{\text{t}}\]

\[\varepsilon = \dot{\varepsilon}_{\text{f}} \left(1 - \exp\left(-52(\dot{\varepsilon}_{\text{crit}})^{0.5}\right)\right) + \dot{\varepsilon}_{\text{t}}\]

\[\dot{\varepsilon}_{\text{f}} = 2.16 \times 10^{-5}(\dot{\varepsilon}_{\text{crit}})^{0.109}\]

\[\dot{\varepsilon}_{\text{t}} = 1.57 \times 10^{10}(E/T)\left\{\sinh(1.13 \times 10^8 \sigma/E)\right\}^{1.1} \cdot \exp\left(-2.72 \times 10^8/R\right).\]

(13)

Attention should be paid that the stress

Figure 10 is the comparison of the experimental data with the predicted creep curves at 633 K, which shows the good applicability of Eqs. (13) for the prediction of the thermal creep behavior of Zircaloy cladding. The correlation coefficient between the experimental data and the predicted values is 0.969.

Attention should be paid that the stress
applied to the tube material under the constant internal pressure has a radial stress distribution, besides it varies with time. Odqvist et al. showed that even a material which has only the steady-state creep stage shows a transient deformation so called "static primary creep" under the internal pressure. It is presumed that these facts may make the primary creep of Zircaloy cladding under the internal pressure look complicated. But the simple treatment of the primary creep by Eq. (12) does not introduce a large error. In fact, the error (standard deviation) is estimated to be 7.6 x 10^{-4} in the primary creep region where creep strain is less than 0.01.

Stehle et al. measured creep loci for Zircaloy cladding at 593~673 K and found that the creep anisotropy was considerably low compared to short time (yield) anisotropy. They showed that Hill's equation for generalized stress could be applied well for the creep behavior but that different anisotropic parameters should be used in the four quadrants of the creep locus. In this study, all the creep tests were conducted under fixed stress ratio; $\varepsilon_\theta/\varepsilon_a = 2.0$ (where $\varepsilon_\theta$ and $\varepsilon_a$ are hoop and axial stress, respectively). The application of the results of this study to other stress ratios in the first quadrant presumes to be possible without large error by converting the creep equations for generalized stress. But for other quadrants, further investigations are necessary to determine anisotropic parameters.

As to in-reactor creep of Zircaloy, it is well known that the creep rate is accelerated by neutron irradiation. The apparent activation energy for the in-reactor creep is reported to be ~10,000 cal/mol (~4.2 x 10^4 J/mol) in "low" (<623 K) temperature range and ~60,000 cal/mol (~2.5 x 10^5 J/mol) in "high" (>623 K) temperature range, which suggests that the thermal activation process is the dominant controlling step in the high temperature range. In fact, Fidleris showed that the in-reactor creep rate of cold-worked Zircaloy-2 at 14.1 kg/mm² (138 MPa) stress was equivalent to the thermal (out-of-reactor) creep rate at temperature higher than 623 K. Therefore, it is presumed that the results of this study can be applied to the in-reactor creep of Zircaloy tubings in the "high" (>623 K) temperature range. But in "low" (<623 K) temperature range, effects of neutron irradiation shall be considered.

**VI. CONCLUSION**

Creep characteristics of Zircaloy-4 stress-
releaved cladding under internal pressure were studied. Creep tests were conducted under 21 conditions chosen from the temperature range of 603~693 K and the hoop stress of 49~314 MPa. The maximum accumulated test period was 3,000 h. The test results indicated that:

1. The diametric creep deformation proceeded uniformly up to more than 10% of strain.
2. The axial elongation was so small in comparison with the diametric strain as to be regarded as zero.
3. Creep data were analyzed by separating the primary (transient) and the secondary (steady-state) creep, based on Dorn’s quasi-theretical model, and the following equations were derived:

\[ \epsilon = \epsilon_s (1 - \exp(-52(\dot{\epsilon}_s t)^{0.5})) + \dot{\epsilon}_s t \]
\[ \dot{\epsilon}_s = 2.16 \times 10^{-2} (\dot{\epsilon}_{trans})^{0.109} \]
\[ \dot{\epsilon}_s = 1.57 \times 10^{19} (E/T) \]
\[ \cdot \sinh(1.13 \times 10^{-3} \sigma/E) \]
\[ \cdot \exp(-2.72 \times 10^7 /RT) \]

4. The apparent activation energy of the steady-state creep, which was \( 2.72 \times 10^5 \) J/mol, was in good agreement with these of self-diffusion of Zr in Zr-Sn alloys and suggested the self-diffusion was the rate controlling step.
5. The complexity of the time dependency of the primary creep might result from the time variation of the stress distribution in wall thickness but the simple treatment didn’t introduce large error in the prediction.

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