In order to investigate irradiation effects on nodular corrosion resistance of Zircaloy-4, an out-of-pile corrosion test was conducted using Zircaloy-4 specimens cut from the channel box of a fuel assembly irradiated in the BWR (Monticello reactor) up to the neutron fluence of $1.53 \times 10^{26}$ n/m² ($E > 1$ MeV). The corrosion test was carried out in high pressure steam of 10.3 MPa at 783 K for 24 h. No nodules appeared on the specimens cut from welded areas of the channel box and nodular corrosion resistance tended to be better with increasing neutron fluence. Microstructural evolution in the form of irradiation-induced release of Fe atoms from Zr(Fe, Cr)₂ type precipitates was detected by an analytical electron microscope. It was found that the higher the concentration of dissolved Fe and Cr in the grains of Zircaloy-4, the better the nodular corrosion resistance.

**KEYWORDS:** out-of-pile test, nodular corrosion, corrosion resistance, Zircaloy-4, precipitation, neutron beams, irradiation, neutron fluence, irradiation-induced release, microstructure, BWR type reactors, amorphization

**I. INTRODUCTION**

Nodular corrosion has often been observed on surfaces of Zircaloy fuel assembly components irradiated in BWRs. It was reported that the growth rate of nodular corrosion decreased with increasing burnup or irradiation time. An out-of-pile corrosion test of neutron irradiated Zircaloy-2 conducted by one of the authors revealed that neutron irradiation improved the nodular corrosion resistance. This tendency was considered to be due to the microstructural evolution of Zircaloy-2 during neutron irradiation. This microstructural evolution was observed by an analytical electron microscope (AEM) and the results indicated that Fe atoms were released from Zr(Fe, Cr)₂ type precipitates, while Fe and Ni atoms were released from Zr₅(Fe, Ni) type precipitates. This suggested that a uniform distribution and/or high concentration of dissolved Fe, Cr and Ni in the grains of Zircaloy-2 improved the nodular corrosion resistance.

In general, dissolved Fe, Cr and Ni in the grains of unirradiated Zircaloy are seldom detected because they are less than the detection limit of energy dispersive X-ray spectroscopy (EDS). They can, however, be detected in neutron irradiated Zircaloy, since their irradiation-induced release from the precipitates could increase the amounts in the grains. In the present work, an out-of-pile corrosion test and AEM investigation were conducted using neutron irradiated Zircaloy-4 to establish a correlation between nodular corrosion resistance and concentration of dissolved Fe and Cr in the grains.
the very small amounts of dissolved Fe and Cr in the matrix, calibration curves for very small amounts of alloying elements in Zircaloy(4) were used.

II. EXPERIMENTAL

1. Specimens

Irradiated specimens for the corrosion test and AEM observations were cut from a Zircaloy-4 channel box used in the Monticello reactor. To get recrystallization, the Zircaloy-4 channel box was annealed (typically at 850 K for 2~3 h). The irradiation temperature was 561±5 K. The accumulated average burnup of the channel box was 59.9 GWd/t and maximum fast neutron fluence was $1.53 \times 10^{26} \text{n/m}^2$ ($E>1 \text{MeV}$). The average fast neutron flux at the maximum fluence region was about $4 \times 10^{17} \text{n/m}^2 \text{s}$. The test specimens were taken from four axial locations and two of them were from welded areas. As a reference, unirradiated, recrystallized Zircaloy-4 was tested simultaneously. Microstructural evolution of some specimens was investigated by AEM.

The designation and fast neutron fluence of each specimen are shown in Table 1.

<table>
<thead>
<tr>
<th>Specimen designation</th>
<th>Fast neutron fluence ($\times 10^{26} \text{n/m}^2$; $E&gt;1 \text{MeV}$)</th>
<th>Weight gain (mg/dm$^2$)</th>
<th>Maximum oxide thickness (μm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>U-1</td>
<td>0</td>
<td>2.372</td>
<td>360</td>
</tr>
<tr>
<td>U-2</td>
<td>0</td>
<td>2.457</td>
<td>—</td>
</tr>
<tr>
<td>NL-1</td>
<td>0.50</td>
<td>2.279</td>
<td>240</td>
</tr>
<tr>
<td>NL-2</td>
<td>0.50</td>
<td>2.679</td>
<td>—</td>
</tr>
<tr>
<td>NH-1</td>
<td>1.44</td>
<td>64</td>
<td>16</td>
</tr>
<tr>
<td>NH-2</td>
<td>1.44</td>
<td>63</td>
<td>—</td>
</tr>
<tr>
<td>WL-1</td>
<td>0.71</td>
<td>433</td>
<td>160</td>
</tr>
<tr>
<td>WH-1</td>
<td>1.53</td>
<td>56</td>
<td>4</td>
</tr>
</tbody>
</table>

Table 1: Corrosion test results

2. Out-of-pile Corrosion Test

The out-of-pile corrosion test was conducted in high pressure steam of 10.3 MPa at 783 K for 24 h in order to investigate the nodular corrosion resistance. Test specimens were about 20×10×2 mm. Their surfaces were mechanically polished to remove oxide layers produced during irradiation and then chemically polished in 5% HF/45% HNO$_3$/50% H$_2$O solution to remove the mechanically deformed layers. Unirradiated specimens were also mechanically and chemically polished to get the same surface condition as the irradiated ones.

All specimens were weighed before and after the corrosion test, and each weight gain was calculated. After the corrosion test, surface appearances of all specimens were investigated, while for selected specimens, oxide layer thicknesses were measured with an optical microscope.

3. Analytical Electron Microscopy (AEM)

Thin foils for AEM were prepared by the twin-jet polishing technique with a 25% HClO$_4$ /75% C$_2$H$_5$OH solution. Typical polishing conditions were 15 V and 223 K. Microstructural evolution was observed by a transmission electron microscope (TEM; Hitachi H-800) and analyzed by EDS (Horiba EMAX-3600).

Intermetallic precipitates in Zircaloy-4 were analyzed by point analyses at the center and boundary. Some characteristic X-rays from Zr in the matrix was also counted during these analyses, because some precipitates were thinner than the thickness of the thin foil. In order to estimate the movement of Fe atoms during neutron irradiation, the fractional concentration of Fe to Fe+Cr($X_{Fe}$) in Zr(Fe, Cr)$_4$ type intermetallic precipitates was calculated using the following equations:

$$X_{Fe,c}=C_{Fe,c}/(C_{Fe,c}+C_{Cr,c})$$
or \[ X_{Fe,b} = \frac{C_{Fe,b}}{C_{Fe,b} + C_{Cr,b}} \]  
where \( C_{Fe} \) and \( C_{Cr} \) are the atomic concentrations of Fe and Cr. Choice of \( X_{Fe} \) was based on the very low concentrations of dissolved Fe and Cr in the matrix, so that \( X_{Fe} \) is scarcely affected by the matrix. Subscripts \( c \) and \( b \) denote the center and boundary of the precipitate, respectively. Then, the ratio \( X_{Fe,b/c} \) was defined as

\[ X_{Fe,b/c} = \frac{X_{Fe,b}}{X_{Fe,c}} \]

The concentrations of alloying elements dissolved in the Zircaloy-4 matrix were also analyzed by EDS. Calibration curves for very small amounts of alloying elements in Zircaloy matrix were used in the analysis. The electron probe was scanned in a square of 100 nm x 100 nm until the integral counts of the Zr-K\(_\alpha\) characteristic X-ray reached 1 million. Detection limits of Sn, Fe and Cr were 0.21, 0.06 and 0.04 at\%, respectively.

III. RESULTS

1. Corrosion Test

Weight gains of tested specimens are summarized in Table 1 and their typical surface appearances are shown in Photo. 1. Relatively large oxide nodules were formed on U specimens. They almost covered the surfaces, but black oxide was also observed on some areas. In general, black oxide, which is thin and dense, is first produced on the surface of Zircaloy. Then, it becomes a thick, porous, white oxide. Small nodules covered all surfaces of NL. On the surfaces of NH, only black oxide appeared, except for white nodules which appeared on the specimen number marking. For WL, no nodules were observed on the welded areas, although white oxide was observed on non-welded areas. By contrast, WH was covered by black oxide.

Weight gains of welded specimens were the average of both welded and non-welded areas and it was difficult to distinguish their relative contributions. Furthermore, U and NL had a thick oxide and even exfoliation of the oxide layer was observed, so that their weight gains were less accurate. Therefore, to evaluate the corrosion resistance, maximum thickness of the oxide layers was measured with an optical microscope.

Typical cross-sectional micrographs of each specimen are shown in Photo. 2. The maximum oxide thickness was measured on these micrographs and the data are summarized in Table 1. A thick, uniform oxide layer (Photo. 2-A), large nodules (Photo. 2-B), and a thin, uniform oxide layer (Photo. 2-C) were observed on U. The maximum thickness was about 360 \( \mu \text{m} \), while the average thickness of the thin, uniform oxide was 2 \( \mu \text{m} \). Only a thick, almost uniform oxide layer (Photo. 2-D) was revealed on NL. This oxide layer might be produced by coalescence of nodules. The maximum thickness was about 240 \( \mu \text{m} \). A thin, uniform oxide layer (Photo. 2-E) covered NH. The maximum thickness was about 16 \( \mu \text{m} \). Although a thick, uniform oxide layer (Photo. 2-F) and large nodules (Photo. 2-G) were observed on the non-welded areas of WL, only a thin, uniform oxide (Photo. 2-H) was observed on the welded area of the same specimen.
The maximum thicknesses were 160 \( \mu m \) for the non-welded and 4 \( \mu m \) for the welded areas. For WH, the difference in thickness between welded (Photo. 2-I) and non-welded areas (Photo. 2-J) was not distinct and both oxides were thin and uniform. Their maximum thicknesses were 6 \( \mu m \) for non-welded and 4 \( \mu m \) for welded, areas.

The maximum thicknesses of the oxide layer are plotted as a function of fast neutron fluence in Fig. 1. The result clearly indicates that the maximum oxide thickness decreases with increasing fast neutron fluence for non-welded areas. This fact suggests that fast neutron irradiation improves the nodular corrosion resistance of Zircaloy-4. A similar effect by fast neutron irradiation on nodular corrosion resistance has already been reported for Zircaloy-2\(^{(4)}\). However, the nodular corrosion resistance of Zircaloy-2 was improved at a very low fluence (ca. \( 2 \times 10^{24} n/m^2 \)). Resistance of Zircaloy-4 was improved at higher fluences and above \( 1.4 \times 10^{26} n/m^2 \) no nodules were formed. The maximum oxide thicknesses of the welded areas were smaller at higher fluences.
than those of non-welded areas and did not depend on fast neutron fluence. Cheng & Adamson reported that welded and heat-affected zones of Zircaloy-4 irradiated in BWRs were more resistant to nodular corrosion than the α-annealed base metal. The present result is in agreement with this.

2. Analyses of Intermetallic Precipitates

The corrosion resistance change observed in this study was attributed to the microstructural change of Zircaloy-4, especially the distributional change of intermetallic precipitates and/or alloying elements. Therefore, the microstructures of unirradiated and irradiated Zircaloy-4, including the welded areas, were investigated by AEM.

(1) U specimens

The hcp-Zr(Fe, Cr) type precipitates were detected in recrystallized Zircaloy-4 and their average size was about 0.2~0.3 μm. The following results were obtained by EDS analyses at the center and boundary of the precipitates: $X_{Fe,c} = 0.63 \pm 0.02$, $X_{Fe,b} = 0.64 \pm 0.03$, hence $X_{Fe,b,c} = 1.01 \pm 0.03$. These indicated that the profiles of the compositional ratio of Fe to Fe+Cr in Zr(Fe, Cr) type precipitates in the unirradiated Zircaloy-4 were uniform.

A typical microstructure of the unirradiated, welded Zircaloy-4 is shown in Photo. 3. The welded region showed lamellar structure and small precipitates on the grain boundary.

(2) NL Specimens

The hcp-Zr(Fe, Cr) type precipitates were also observed in NL specimens. They showed amorphization from their periphery and selective irradiation-induced release of Fe atoms as reported by other authors. A typical TEM image and an electron diffraction pattern of Zr(Fe, Cr) type precipitate are shown in Fig. 2. The EDS spectra at the center and boundary of the precipitate are shown there. Figure 2(A) is the bright field image of the precipitate, in which the outer region exhibited different contrast from the center region. A halo ring and diffraction patterns of hcp-Zr(Fe, Cr) and hcp-Zr were observed in a selected area diffraction pattern (SAD pattern) of the precipitate shown in Fig. 2(B). The diffraction spots from hcp-Zr(Fe, Cr) are linked by lines in Fig. 2(B). These photos indicated partial amorphization of the precipitate. The EDS spectra at the center and boundary, as shown in Fig. 2(A), are shown in Figs. 2(C) and (D), respectively. The values of $X_{Fe}$ decreased in comparison with those in unirradiated samples; in particular $X_{Fe,b}$ showed a remarkable decrease. The compositional ratio of the boundary to center $X_{Fe,b,c}$ was about 0.5 for relatively large precipitates. These results indicated selective Fe release from the outer region of the precipitate.

(3) NH Specimens

All of the precipitates observed in NH specimens showed complete amorphization. An example of total amorphization of a relatively large precipitate is shown in Fig. 3(A) as the dark field image of the precipitate. The SAD pattern of this precipitate is shown in Fig. 3(B). This pattern shows a halo ring and the diffraction pattern of hcp-Zr. The EDS spectra at the center and boundary are shown in Figs. 3(C) and (D). Both $X_{Fe,b}$ and $X_{Fe,c}$ were much smaller than those in U specimens. But $X_{Fe,b,c}$ was nearly 1. This result indicated that the distribution of Fe and Cr in the precipitate was almost uniform in the high fluence samples since Fe depletion occurred even at the precipitate center.

(4) WL Specimens

Many very small precipitates were observed along the grain boundary or sub-grain boundary in unirradiated welded Zircaloy-4. No precipitates, however, were observed in WL
Fig. 2 TEM image and EDS spectra of Zr(Fe, Cr)₂ type precipitate in low fluence Zircaloy-4 (0.50 x 10²⁶ n/m²; E>1 MeV)

A: Bright field image of partially amorphized precipitate. Arrows b and c indicate the analyzed points. B: Selected area diffraction (SAD) pattern of the precipitate shown in A. The linked spots are diffraction pattern of hcp-Zr(Fe, Cr)₂. This photo also includes the pattern of hcp-Zr. C: EDS spectrum at the center of the precipitate indicated by arrow c in A. D: EDS spectrum at the boundary of the precipitate indicated by arrow b in A.

Fig. 3 TEM image and EDS spectra of Zr(Fe, Cr)₂ type precipitate in high fluence Zircaloy-4 (1.44 x 10²⁶ n/m²; E>1 MeV)

A: Dark field image of totally amorphized precipitate. Arrows b and c indicate the analyzed points. B: SAD pattern of the totally amorphized precipitate shown in A. C: EDS spectrum at the center of the precipitate indicated by arrow c in A. D: EDS spectrum at the boundary of the precipitate indicated by arrow b in A.
specimens. These small precipitates might be completely dissolved into the matrix.

(5) Shortest Path Dependence of $X_{Fe,c}$

The Fe depletion of the precipitate occurred from the surface of the precipitate and proceeded inwards. Therefore, Fe depletion at any point in the precipitate should depend on the distance from the surface. It could be supposed that the ratios of Fe to Fe+Cr at the center of the precipitates $X_{Fe,c}$ depended on the shortest distance from the center to the surface. The center was defined as the farthest point from the precipitate surface. The center was determined using an inscribed circle of the precipitate, as shown in Fig. 4.

Hence, the shortest path (SP) was defined as the shortest distance from the center to the surface, i.e. the radius of the inscribed circle. The shortest path dependence of $X_{Fe,c}$ is shown in Fig. 5.

For NL specimens, $X_{Fe,c}$ did not depend on the shortest path. The average and standard deviation of $X_{Fe,c}$ were 0.63 and 0.02.

For NH specimens, $X_{Fe,c}$ also had large scattering, but there was a slight dependence on the shortest path. The values of $X_{Fe,c}$ were much smaller than those of U and NL. Specimens NL and NH were taken from different axial positions of the channel box. Their irradiation time and temperature (coolant temperature) were the same, so that Fe depletion depended on fast neutron flux. The higher fast neutron flux depleted more Fe from the precipitate.

The ratios of Fe to Fe+Cr at the boundary of the precipitates $X_{Fe,b}$ did not show clear dependence on the shortest path, although $X_{Fe,b}$ in the irradiated specimens increased slightly with increasing shortest path. The averages of $X_{Fe,b}$ for each specimen were $0.64 \pm 0.03$ for U, $0.32 \pm 0.09$ for NL, $0.15 \pm 0.09$ for NH. This also showed that Fe depletion was accelerated by fast neutron flux.

3. Alloying Element Distribution in the Matrix

Examinations of the precipitates in neutron irradiated Zircaloy-4 showed that mainly Fe atoms moved from the precipitates into the matrix due to fast neutron irradiation in BWRs. To investigate the distributions of dissolved alloying elements in the Zircaloy-4 matrix, the element concentrations were measured by EDS. The nominal concentrations of Fe and Cr in Zircaloy-4 were less than 0.24 wt% (ca. 0.39 at%) and most of the Fe and Cr atoms were present in the precipitate. Their concentrations in the matrix were too small to detect by a normal EDS measurement technique. To detect these very small amounts of alloying elements, EDS measurements were made until the integral counts of the Zr-K$_a$ characteristic X-ray reached 1 mil-
lion. Calibration curves for measurement of very small amounts of alloying elements in Zircaloy have been established(4) and they were used here. Detection limits of Fe and Cr were 0.06 and 0.04 at%, respectively.

Figures 6 and 7 show TEM images of analyzed regions and the distribution profiles of the alloying elements in U and NL. The analyzed regions included precipitates, and the origin of the distance was the interface between the precipitate and matrix. The sign of the distance was positive in the matrix and negative in the precipitate. In both figures, Cr atoms were detected in the matrix near the precipitate, but they could not be detected far from the precipitate. Iron atoms were detected in the matrix farther from the precipitate in NL than in U. This suggested that irradiation-induced release of Fe atoms occurred in the former. However, Fe concentration in the matrix far from the precipitate was too low to detect.

Figure 8 shows the TEM image of the
analyzed area and concentration profiles of alloying elements in NH specimens. The precipitate was well amorphized in the analyzed area as shown in Fig. 3. In the precipitate, the Fe concentration was much lower than those in U and NL. Iron atoms could be detected in the matrix away from the precipitate and the concentration in the matrix was almost constant.

Specimens WL showed the same lamellar structure as welded regions of U specimens as shown in Photo. 3, but had no precipitates as shown in Fig. 9. EDS analyses were conducted across the grains and the results are given in Fig. 9. These profiles showed high concentrations of Fe and Cr atoms at the grain boundary. This result would correspond with the profiles of the small precipitates in welded regions of U specimens as shown in Photo. 3. These small precipitates might be completely dissolved by neutron irradiation. Then, high concentrations of Fe and Cr atoms would be established in the vicinity of the grain boundary. Figure 9 also showed that both Fe and Cr were detectable in the matrix and Fe had different concentrations in each grain. This might reflect the unisotropic mobilities of interstitial atoms in the hcp-Zr matrix; i.e. interstitial atoms could diffuse faster along the c-axis than in the perpendicular direction\(^{(8)(9)}\).

The average concentrations of alloying elements at grain boundaries and in grains away from the precipitate are summarized in Table 2. Iron and Cr were not detected in the matrix of U, since their solubility limits are less than EDS detection limits. In NL, Fe atoms were detected only at the grain boundary and Cr atoms were not detected. In NH, however, Fe atoms were detected, even in the grains, but Cr atoms were not. For WL, both Fe and Cr atoms were detected.

![Image A. TEM IMAGE](image)

![Image B. CONCENTRATION DISTRIBUTION](image)

**Table 2.** Alloying element concentrations in Zircaloy-4 matrix

<table>
<thead>
<tr>
<th>Specimen designation</th>
<th>Fast neutron fluence ( \times 10^{20} \text{n/m}^2; E&gt;1 \text{MeV} )</th>
<th>Alloying element concentration (at%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>in the grain</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Sn</td>
</tr>
<tr>
<td>U</td>
<td>0</td>
<td>1.53</td>
</tr>
<tr>
<td></td>
<td>0.15</td>
<td>—</td>
</tr>
<tr>
<td>NL</td>
<td>0.50</td>
<td>1.39</td>
</tr>
<tr>
<td></td>
<td>0.05</td>
<td>—</td>
</tr>
<tr>
<td>NH</td>
<td>1.44</td>
<td>1.32</td>
</tr>
<tr>
<td></td>
<td>0.15</td>
<td>0.13</td>
</tr>
<tr>
<td>WL</td>
<td>0.71</td>
<td>1.30</td>
</tr>
<tr>
<td></td>
<td>0.05</td>
<td>0.24</td>
</tr>
</tbody>
</table>

*Upper values show average concentrations and lower standard deviations.
"—" stands for "not detected".*
at the grain boundary and in the grain. These results suggested that neutron irradiation increased the concentrations of dissolved alloying elements. Welding produced smaller precipitates, which more easily dissolved during neutron irradiation.

IV. DISCUSSION

It was found in this study that nodular corrosion resistance of Zircaloy-4 was improved by neutron irradiation. A synergistic effect of welding and neutron irradiation more drastically improved it. Furthermore, neutron irradiation increased the concentration of dissolved alloying elements in the matrix and neutron irradiation on the welded region completely dissolved the precipitates. It was already known that alloying elements, such as Fe and Cr, improved the corrosion resistance of Zr. Neutron irradiation and welding changed the distribution and/or the state of alloying elements, such as dissolution in the matrix or precipitate formation. Therefore, the nodular corrosion resistance change of Zircaloy-4 due to neutron irradiation and welding were attributable to the change of distribution and/or the state of the alloying elements. Some reports have shown that the nodular corrosion resistance of Zircaloy depended on the average size or number density of the precipitates. But others have found some exceptions in which the average size and number density of the precipitates could not completely explain the nodular corrosion resistance. In this study, it was also found that welded and neutron irradiated Zircaloy-4 in which no precipitates were detected showed the best nodular corrosion resistance. This indicated that nodular corrosion resistance did not directly depend on the morphology of the precipitates.

Cheng & Adamson proposed a nodular corrosion model which was based on the different corrosion resistance between ZrO$_2$ and ZrO$_{2-x}$, in which deviation from stoichiometry was produced by substituting Fe and Cr for Zr. The valences of Fe and Cr are smaller than Zr. Based on their model, higher concentrations of dissolved Fe and Cr atoms in the metal produce a more resistant oxide film, because they easily occupy the substitutional site in the oxide. Therefore the nodular corrosion resistance of Zircaloy-4 is improved. Figure 10 correlates the maximum oxide thickness formed in the out-of-pile nodular corrosion test and the concentration of dissolved Fe and Cr atoms in the grains away from the precipitate. It could be concluded that nodular corrosion resistance of Zircaloy-4 depended on the concentration of dissolved Fe and Cr atoms in the grain.

![Figure 10](image-url)

We previously showed that nodular corrosion resistance of Zircaloy-2 was drastically improved due to very small fast neutron fluence (ca. $2 \times 10^{21} n/m^2$) as shown in Fig. 11. Nodular corrosion resistance of Zircaloy-4 was gradually improved with increasing fast neutron fluence. This difference in fast neutron fluence dependency between Zircaloy-2 and Zircaloy-4 might be due to the difference in the irradiation-induced release rate of alloying elements between them. For Zircaloy-2, there are two types of intermetallic precipitates, i.e. Zr(Fe, Cr)$_3$ and Zr$_2$(Fe, Ni). It was seen that the Zr$_2$(Fe, Ni) type precipitate was more rapidly dissolved than Zr(Fe, Cr)$_3$ type precipitate under BWR irradiation conditions. Therefore, concentrations of dissolved Fe and Ni atoms in the matrix increased more rapidly.
in Zircaloy-2 than Zircaloy-4. Consequently, the nodular corrosion resistance of Zircaloy-2 was improved faster than that of Zircaloy-4 during neutron irradiation.

The observations of nodular corrosion on Zircaloy irradiated in BWRs showed that the growth rate of nodular corrosion decreases with burnup\(^{(1)}\) or irradiation time\(^{(2)}\). The main cause of this tendency was considered to be the improvement of the nodular corrosion resistance of Zircaloy during neutron irradiation. At the beginning of irradiation in a BWR, the concentrations of dissolved Fe, Cr and Ni in the grains are not high enough to prevent nodular corrosion. Nodular corrosion appears and grows during irradiation. But as the concentrations of dissolved Fe, Cr and Ni increase with irradiation time, the nodular corrosion resistance of Zircaloy is gradually improved. As a consequence, the growth rate of nodular corrosion decreases with irradiation time.

V. CONCLUSIONS

Neutron irradiation effects on nodular corrosion resistance and microstructural evolution of Zircaloy-4 were investigated by an out-of-pile corrosion test of neutron irradiated Zircaloy-4 and analytical electron microscopy. The following results were obtained:

(1) Neutron irradiation improved nodular corrosion resistance of Zircaloy-4. Neutron irradiation on welded regions raised the nodular corrosion resistance more than that irradiation on non-welded area.

(2) Selective irradiation-induced release of Fe atoms from the precipitate and amorphization of the precipitate occurred during neutron irradiation.

(3) No precipitate was detected in the welded region of neutron irradiated Zircaloy-4.

(4) A strong correlation was found between the nodular corrosion resistance and concentrations of dissolved Fe and Cr in the grains.

ACKNOWLEDGMENT

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