Characterization of Pellet-Clad Mechanical Interaction in a failed spent fuel rod

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1. Introduction

A failed spent fuel rod with 53,000 MWd/tU from a nuclear power plant was characterized, and the fission products and oxygen layer in the Pellet-Clad Mechanical Interaction region were observed using an EPMA (Electron Probe Micro-Analyzer, CAMECA SX-50R). A sound fuel rod burned under similar conditions was used to compare and analyze the results of the failed fuel rod. The formation of a bonding layer is an important phenomenon in the mechanical interaction between fuel pellet and cladding for highly burned fuels. The bonding layer limits the independent movement of fuel pellets and cladding, and thus generates higher stress on the cladding during transient reactor operation. EPMA techniques offer the possibility of identifying and analyzing phases and segregations in spent PWR fuel. Although phases and segregations are small in terms of the amount expected to be present in background radiation, they nevertheless present a significant analytical challenge. The gap between the cladding layer and the bonding layer in the failed fuel was about 20 μm. The existence of a bonding layer with a size of 20~30 μm beyond the PCMI gap was confirmed. The cladding inner surface corrosion and its resulting fuel-cladding bonding were investigated using an EPMA.

2. Sample and Discussion

2.1 Sample preparation

A thin diamond wheel was used to cut samples from a PWR failed spent fuel rod with 56,000 MWd/tU, that had been withdrawn from a nuclear power plant and cooled down for two years. In addition, a 53,750 MWd/tU burned sound fuel rod was cut with a diamond wheel for comparison with the results of the failed fuel specimen. The work for cutting and preparing the fuel rod specimens preceded in a hot cell space designed to enable the operation of highly radioactive materials.

The samples were embedded in epoxy resin and polished with diamond grinding disks of successively finer grain size, and finished with a diamond paste of 1 μm as the final stage. Before mounting each sample in the EPMA, the samples were coated with carbon to prevent charging. The carbon-coated specimens were mounted in a holder together with the X-ray standard. The EPMA was performed using a CAMECA SX-50R equipped with a two wavelength dispersive X-ray spectrometer shielded with tungsten.

In the EPMA, the radiation activity allowed was up to 3.7 x 1010 Bq. The specimen holder as a part of the equipment was shielded from radiation leakage, and wavelength dispersive spectroscopy count windows containing lead and tungsten were used for analysis of the irradiated nuclear fuel [1].

The analysis was performed with a beam current of 50 nA at an accelerating voltage of 25 kV, in order to give a reasonable peak-to-background ratio for an active specimen. It was operated so that an electron beam was focused exactly at the center of the fuel-clad gap and crud layer. At the same time, tiny vibrations were excluded during measurement by turning off the stage motor while measuring. The electron beam was in fixed mode with 1 μm beam size.

The γ-activity was maintained within the tolerances specified above. Although a specimen has to be maintained at a sufficiently small size, its volume can be managed with a manipulator in a hot cell. After cutting the specimens into a manageable size with the manipulator, the specimens were hot- mounted with a conducting resin at 150 ℃ and 0.6 MPa. An overly thin specimen is apt to break during hot mounting, and thus a specimen was cut to about 5 mm of its depth at first, and finally a specimen of 2.5 x 5 x 1.5 mm3 was fabricated by repeating the mounting and cutting processes several times to decrease the radiation level by using a manipulator. The specimen was subsequently polished in a conventional manner with γ -monitoring until the activity was considered to be low enough to handle it.

2.2. Discussion

Figure 1 shows the PCMI region oxide layer profile of the failed fuel and normal fuel rod. The normal fuel rod specimen was extracted at a height of 2,957 mm from the bottom, which was similar to the failed fuel rod specimen location. The average design burn-up of the fuel rod was 53, 750 MWd/tU, having a burning history similar to that of the failed fuel rod. As shown in Fig. 1A, it can be seen that the oxide layer represents 10 μm of the boundary of the cladding, and 35 μm of the region outside the cladding area. In the profile of Zr and O in Fig. 1B, an oxide layer with a size of 8 μm is observed in the cladding boundary region. This result is in contrast to Fig. 1A.

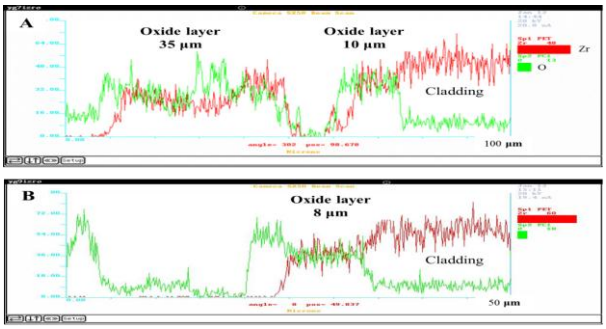

Fig.1 Profiles of Zr, and O in the failed fuel and sound fuel rod cladding PCMI region.

Figure 2 shows the SEM images and X-ray image mapping at the same positions as shown in the profile of Fig.1A. In the SEM image of Fig. 2, there is a compound with a thick thickness of about 20~30 μm after the cladding and bound resin region. If a gap is initially present in the rod, a large pellet strain is required to produce mechanical interaction with the cladding, which will take place particularly in regions next to the pellets ends. The fuel rod acquires in that case the aspect of a cane (bamboo effect). This bamboo effect compound is known to cause PCMI, if the pellet deformation is sufficiently large as a bonding phenomenon due to UO2/Zircaloy interaction. This phenomenon is more thermodynamically unstable than the fuel, so chemical interaction between the fuel and the cladding may occur when the fuel rod is exposed to high temperature [2].

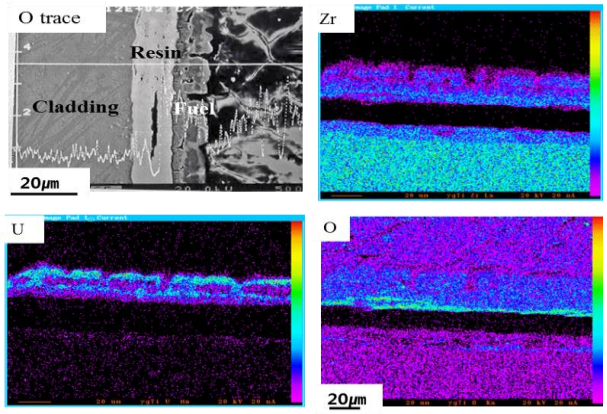

Fig. 2 SEM image of PCMI in the failed fuel rod and X-ray image mapping of O, Zr, and U.

According to data from Koji MAEDA [3], the formation process of the bonding layer as described above can be explained as follows:

Oxidation of Zr (Monoclinic ZrO2, Stable below 1170 ℃ fission damage in ZrO2) Phase transformation of ZrO2 (Monoclinic to cubic, Gap closure(cladding creep down, fuel swelling) Strong contact of cubic ZrO2 film and UO2 pellet - Mutual diffusion of U and Zr Formation of (U,Zr)O2 solid solution Bonding layer.

The X-ray image mapping of U, Zr, and O in Fig. 2 shows that the bonding layer thickness of (U,Zr)O is estimated to be 20~30 μm. In Figure 2, the gap between the cladding layer and the bonding layer is estimated to be about 20 μm. What is unusual is that uranium does not exist at the cladding interface at all. On the contrary, zirconium exists in the bonding layer of the (U,Zr)O compound beyond the PCI gap of 20 μm, and oxygen also shows a similar tendency to zirconium. In many papers related to PCMI, reference is often made to the bonding layer, but it is difficult to observe the SEM image of the bamboo effect phenomenon. In this study, however, we have identified an SEM image demonstrating the bamboo effect phenomenon as shown in Fig. 2. This was observed only in failed nuclear fuel rods.

In a normally burned fuel rod, it can be seen that the bonding layer of U exists not only in a thickness of several micrometers at the cladding interface but also in a constant band on both sides of the PCMI gap. It is assumed that the bonding layer of U is generated by the pellet-clad mechanical interaction due to the power bump during high-combustion for long periods.

Figure 3 shows the results of quantitative analysis of the distribution of Zr, U, Sn and O. The ratio of Zr/O, and average values from 8 to 17 in the axial direction are shown in Fig 3. Zr is 37.62 at% and O is 61.22 at%. Its chemical formula can be written as ZRO2. The result of the analysis point, where the gap between the cladding layer is very narrow. Unfortunately, in Figure 2, quantitative analysis of the entire region from cladding to the bonding layer representing the bamboo effect was not analyzed.

Some researchers have confirmed that Zr2O, ZrO2, Zr2O3, and so on sub-oxides are formed on the surface depending on the oxygen partial pressure supplied when oxidizing Zr [4]. It is also known that even when Zircaloy-4 cladding is oxidized, low-oxidation oxides such as ZrO2 and Zr2O3 are formed together with ZrO2 [5].

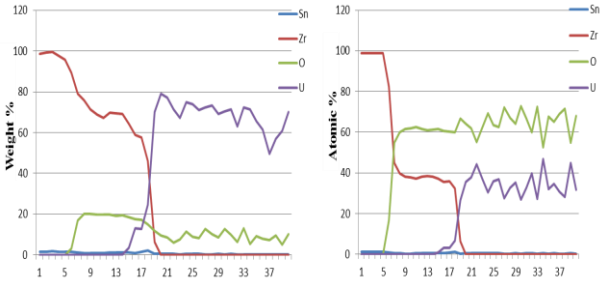

Fig. 3 Quantitative analysis of O, Zr, and U on inside cladding oxide and bonding layer

The composition of U, Zr and O in the bonding layer of Fig. 3 showed that the concentration of Zr was 33.28 wt%, U was 43.31 wt%, and O was 13.98 wt%. The total concentration measured in this region was 90.53 wt%. The weight ratio of this compound to atomic ratio

is as follows: The concentration of U was 12.87 at%, Zr was 25.76 at%, and atomic ratio oxygen was 61.36 at%. Its chemical formula can be written as UZr2O3.

As is well known, it is difficult to measure the quantitative value of oxygen using an EPMA analyzer. The surface condition of the specimen, the composition of the specimen and the conductivity are obstacles to the accurate analysis of the quantitative value of oxygen.

Of course, analysis of oxygen composition in spent fuel is also difficult. This means that it is not easy to analyze the exact composition because the conductivity of the fuel is much lower than that of common metals. The results indicate these data in order to prove that there is no error in the measurement of oxygen.

3. Conclusions

In the failed fuel rod, the oxide layer represented 10 μm of the boundary of the cladding, and 35 μm of the region outside the cladding. By comparison, in the sound fuel rod, Oxide layer size was 8 μm, which was observed in the cladding boundary region.

If the pellet deformation is sufficiently large as a bonding phenomenon due to UO2/Zircaloy interaction, the fuel rod takes on the aspect of a cane (bamboo effect).

Zirconium exists in the bonding layer of the (U,Zr)O compound beyond the PCI gap of 20 μm, and oxygen also shows the tendency of zirconium in the failed fuel rod. In this study, an SEM image demonstrated the bamboo effect phenomenon, but it was only observed in the failed fuel rods. The composition of U, Zr, and O in the bonding layer of the failed fuel was 12.87 at%, 25.76 at%, and 61.36 at%, respectively. Its chemical formula can be written as UZr2O3.

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