# Characteristic of Zircaloy-fuel Mechanical Interaction in Failed Spent PWR Fuel

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## 1. Introduction

In typical PWR fuel rods, the end-of-life internal pressure can be significant because of the initial helium backfill pressure and the added pressure from the release of fission gases. Fuel-clad interaction and the formation of fuel-clad bonding layers with specified chemical, physical and mechanical properties are of importance with regard to the evolution of thermal conductivity as well as in the context of a Pellet-Clad Mechanical Interaction (PCMI). It is also important in the framework of the long-term storage of spent fuel where the phases formed at the fuel-clad boundary are considered to be the first to be leached in the case of a cladding failure [1]. The small volume of material analyzed and the easy of quantification made EPMA the ideal analytical tool with which to study the nuclear industry by providing fundamental information about the behavior of nuclear fuel under extreme irradiation conditions and about the performance of new fuel designed for the in-pile incineration of nuclear waste [2].

## 2. Sample preparation

A thin diamond wheel was used to cut off the samples from a PWR failed spent fuel rod with 53,000 MWd/tU and normal spent fuel rod with 62,000 MWd/tU, which were withdrawn from the nuclear power plant and cooled down for 2 and 4 years, respectively. The samples were embedded in an epoxy resin and polished with diamond grinding disks of successively finer grain size, finishing on cloth with diamond paste of 1 ㎛ as the final stage. Before mounting the sample in the EPMA, the samples were coated with carbon to prevent charging. The carbon-coated specimen was mounted in a holder together with the X-ray standard. The EPMA was performed on a CAMECA SX-50R equipped with a two wavelength dispersive X spectrometer shielded with tungsten.

## 3. Results and discussions

Fig. 1-A shows an SEM image of failed spent fuel with 53,000 Mwd / tU. In the figure, the fuel-clad gap has an empty space of about 10 μm. This is a normal shape observed in spent fuel. At the boundary of the cladding, the fuel portion was observed at a thickness of about 5 to 10 μm, and the thick oxide film was clearly observed in the region corresponding to the fuel portion. Of course, such an oxide film was not observed over the entire region. Performed a quantitative analysis at a distance of 30 μm as indicated by the arrows on the part of the oxide film observed.

Figure 1-B shows the result of a 30 point quantitative analysis for the fission products. The horizontal axis of the figure is the point analyzed at 1 μm intervals, and the vertical axis is the quantitative analysis value. As shown in the figure, the concentration of the fission product from 1 to 17 points hardly appears, but it shows a rapid increase in the concentration until the oxide film thickness of 17 to 23 points is reached. In particular, the concentrations of Cs and Ce are as high as 0.5wt%. It is considered that the fission products in the fuel-clad gap penetrated into the oxide layer.

Figure 1-C shows the results of a quantitative analysis of O, Zr, and U at 53,000 Mwd / tU with failed spent fuel. The zirconium concentration was 42wt% at 15 to 23 points, which is the oxide layer part, and the concentration of oxygen was 58wt%. It is represented through the formula of ZrO8x. It was confirmed that the concentration of oxygen was more than twice that of the zirconium dioxide layer. This expansion of the oxide layer not only weakens the properties of the cladding but also has the possibility of breakage.

Figure 1-D shows the results of a quantitative analysis of O, Zr, and U with 62,000 Mwd / tU of normal spent fuel. The oxygen concentration at 15 to 25 points is about 15 to 18wt%. At a high burn-up, the oxidation thickness and oxygen concentration in the fuel-clad gap of the spent fuel were much smaller than in Figure 1-C. The concentration distribution of Zr and U at the higher oxygen region is illustrating the general trend for the redistribution in the fuel-clad gap region.

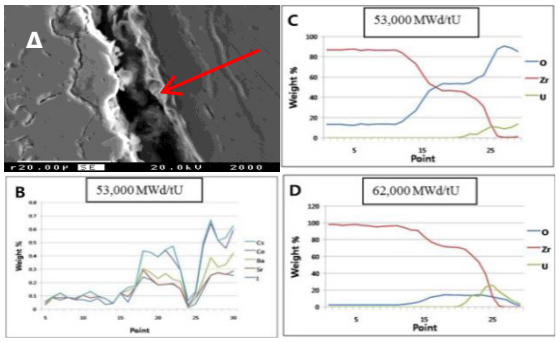


Fig. 1. SEM image of 53,000 MWd/tU and 62,000 MWd/tU fuel rods and quantitative analyses of O, Zr, U and, Cs, Ce, Ba, Sr and I on the marked point.

Fig. 2 illustrates the general trend for the release and cesium redistribution in the fuel-clad gap with 53,000 MWd. The distribution of O, Zr, and U in the figure clearly shows the state at the periphery of the fuel-clad gap. However, the distribution of fission products cannot observe a specific trend. The reason for this is that the concentration of fission product is extremely low, indicating only a general tendency. The distribution of fission products was tested under the same conditions in high burn-up spent nuclear fuel, but no particular trend was observed.

Halogen and iodine are two of the most volatile fission products, and their high release has been reported from early isochronal annealing experiments on irradiated UO2 at various temperatures. The diffusion coefficient of the halogen in single and polycrystalline UO2 during irradiation is two orders of magnitude higher than that of xenon.

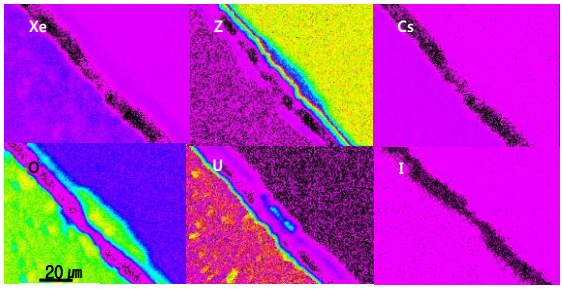


Fig. 2. Image mapping of Xe, Zr, O, U, Cs and I on fuel-clad gap with 53,000 MWd/tU.

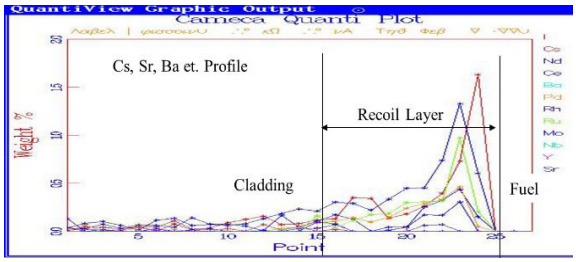


Fig. 3. Distributions of fission products in Recoil layer region with 53,000 MWd/tU.

Fig. 3 shows a qualitative concentration profile along the clad-fuel gap with 53,000 MWd / tU. Zircaloy cladding is known to have a strong affinity for oxygen. The in-reactor corrosion of the cladding typically produces a thin outer layer of oxide 8-40 ㎛ in thickness. Under PWR operating conditions, the oxidation of Zircaloy is suppressed by the time presence of dissolved hydrogen in the primary coolant. However, oxidation rates in air at temperatures of 400℃ can be significantly higher. Thus, at a storage temperature of 400 ℃, the degradation of cladding due to oxidation is found to be for all of the zircaloy cladding, which will be oxidized. During reprocessing of spent fuel elements, the fuel rods are chopped into pieces a few cm in length and the spent fuel content is dissolved in an acid solution. The remaining hulls are contaminated with actinides and fission products [3]. The characterization of the hulls is of interest not only for the safety of waste disposal but also for improving the processing process. The cladding materials remain after the processing of nuclear fuel, generally called hulls [4, 5].

## 4. Conclusion

A failed spent fuel rod with 53,000 MWd/tU and a normal spent fuel rod with 62,000 MWd/tU from a nuclear power plant were characterized to compare and observe the fission products in the fuel-clad gap region using the EPMA. The concentration of fission product shows a rapid increase in the concentration until the oxide film. In particular, the concentrations of Cs and Ce are as high as 0.5wt%. The zirconium concentration was 42wt% at the oxide layer part, and oxygen was 58wt% on 53,000 MWd/tU sample. It is represented through the formula of ZrO8x. It was confirmed that the concentration of oxygen was more than twice that of the zirconium dioxide. The formation of such an oxide layer causes a fatal defect in the integrity of the cladding tube, which can be considered as a fear of damage in the past. On the other hand, it was confirmed that the oxygen concentration of the oxidized layer of normal high burn-up spent fuel was about 15-18wt%.

## REFERENCE

[1] S. Van den berghe, A. Leenaers.., Seminar NEA proceedings on the PCI in Water Reactor Fuels, Aix-en-provance, France, 266, 9-11 March (2004).

[2] Clive Walker “Electron Probe Microanalysis of irradiated nuclear fuel: an overview” J. Analytical Atomic Spectrometry 1999, 14, 447-452The MTI Mechanical tester for JEOL-7600: (Info from Jan-Fong on 4-18-2016).

[3] R. Restani, E.T. Aerne et al. “Characterization of PWR Cladding Hulls from Commercial Reprocessing” Technical Report 92-13, NAGRA (1992).

[4] In-Ha Jung, et al. “Investigation of PWR Hull with a View to Downgrade” J. Separation Science and Technology, 41(10), 2006.

[5] Shin J. M. et al. “A state of the art report on the characterization of the spent PWR cladding hull” Technical report. KAERI/AR-630/2002.