Analytical Techniques of CRUD by EPMA

Y.H. Jung[1], S.J. Baik, B.O. Yoo, S.B. Ahn, W.S. Ryu
Irradiated Materials Examination Facility, Korea Atomic Energy Research Institute,
Deokjin-dong, Yuseong-gu, Daejeon, 305-353, Korea
[*]Corresponding author. E-mail : nyhjung@kaeri.re.kr

1. Introduction

Activated corrosion products CRUD (Chalk River Unidentified Deposit) from BWR and PWR reactors deposit primarily on the outer surfaces of fuel rods[1]. The generic term 'crud' has been applied to particulate material deposits on the surface of fuel rods and assemblies. During reactor operations, crud deposits on the outer surface of all fuel rods. Also, it can become detached in cooling water and storage systems[2], causing additional radiation exposure to plant workers. Despite its importance, crud is difficult to characterize by direct analysis.

Data from EPMA (Electron Probe Micro Analyzer, CAMECA SX-50R, France) provided in this study detail information about the ways of crud collection, chemical compositions, and analysis according to the method of extraction of crud samples on tiny areas.

2. Samples and Methods

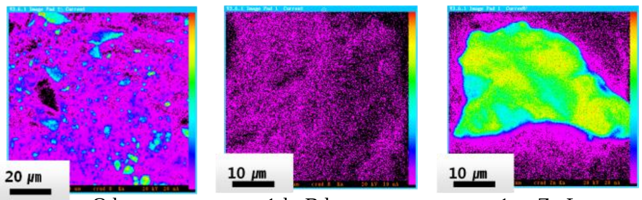
As part of a continuing effort to improve the way of crud sampling, there are a number of ways in order to develop crud collection from irradiated nuclear fuel cladding. An average burn-up of 40,000 MWd/tU for a sampling of hard crud with a steel knife was used in this study.

2.1 Sampling Using a Metal Knife

EPMA image analysis was attempted by gathering crud samples on the surface of the fuel cladding tube using adhesive tape and rubbing paper. Although the specimen was coated with carbon and tried for electron conductivity of the specimen surface, but it was impossible to perform the EPMA analysis, because of dispersion the beam charging from the specimen surface.

Figures 1 shows image analysis of the crud sample scraped by a metal knife. Image analysis was executed concerning oxygen, boron, and zinc by using EPMA. Figure 3 is the test result for the image distribution on oxygen in order to investigate the distribution for the general composition of the sample.

In preparing the sample, the piece of sample, scraped by a metal knife, was carefully put on the adhesive carbon tape in the Groove box and dried. Also there is no way to know the direction and location of the specimen. As can be seen in the figure, it was found that oxygen was distributed unequally as fragments of several ㎛. Distribution of Zr, which is the main material of fuel cladding tube, iron and nickel, which are known as the main elements of the crud, were investigated, but it was not the same as with the oxygen. This means that the distribution of oxygen, shown in the Figure 1, is the compound of crud, or fragments of the oxidized fuel cladding tube were generated by the metal knife.



1-a, O ka 1-b, B ka 1-c, Zn La

Fig. 1. Crud particles x-ray image of O ka, B ka, Zn La by a steel knife in a KAERI-PIEF Hot-cell.

2.2 Sampling Using a cutting fuel cladding tube

Figure 2 was obtained by analyzing a sample prepared for the process by which the fuel cladding tube was cut by a diamond saw, and inner nuclear fuel was removed. Such samples allow not only the exact investigation of thickness and shape of the portion of crud that is attached to the cross-section of the fuel cladding tube, but also provides an overview of the distribution of the crud. Figure 2-c is the image distribution of the iron.

It was found for the figure the thickness and shape were well observed, and the composition gradient was highest according to the location but not with an equally distributed iron. Not only the result of the analysis about oxygen and nickel on the same location, but also the result of the analysis about chromium, manganese, cobalt, and copper that are potential elements which could be made by corrosion or a corroded product from the primary water used at the common power plant as described in the Figure 2-b. Figure 2-b shows the result of the quantitative analysis for the marked point in the Figure 2-c.

In order for the samples to represent the test materials in the crud test, the existence of boron and specific elements should be investigated, first by executing an overall qualitative analysis of the surface of fuel cladding tube (cf. ample transfer distance is 20,000 ㎛). To analyze the constituents of crud for an approximate 8 ㎛ thickness, penetrating the depth of the electrons, that are generated from the analyzing machinery, into samples should be considered, and location and composition of boron and specific element on the crud may be able to be investigated after at least 40 kV of voltage is supplied.

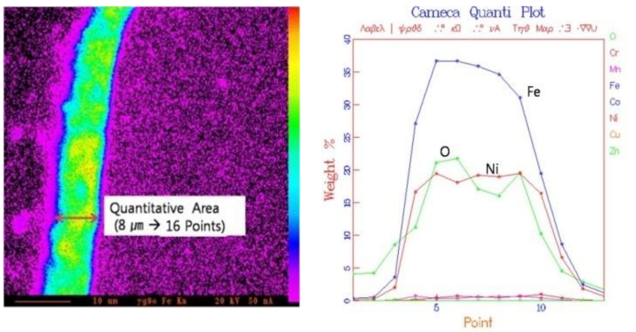


Fig. 2. Crud particles x-ray image of O ka, B ka, Zr La by a by a cross-section using a diamond wheel cutter in a Hot-cell.

2.3 Sampling Using a an Ultra-Sonic System

Figure 3-a shows the hexagonal form of precipitates, in the sample that crud, absorbed on the fuel cladding tube, was shaken off utilizing the ultrasonic system, and Figure 3-b shows the boron crystals. it was found that the composition of these are similar to zirconium, iron, and nickel oxide rather than to Trevorite(NiFe2O4) compound which is the main oxide of crud[3,4].

In the Figure 3-b, the boron composition of some precipitate crystals are relatively uniform and high, but that of some other precipitate crystals are relatively low.

Figure 3-c shows the analysis about iron in the same region. It was found that the composition of iron was not unequal in the area that is not a constituent part of boron, and the result of analysis about oxygen showed that oxygen did not exist in the boron crystal. Table 1 shows the specific area of the result that is obtained by the quantitative analysis of a boron precipitate in the method of electronic beam concentration.

Concentration of heavy elements and light elements were almost zero in most result of quantitative analysis, and sometimes there are areas where the concentration of silicon was detected. However, results in the Table 2 is thought be as a result of the reaction of substances distributed peripherally rather than a chemical composition of the crystal, and also the composition is similar to the constituent composition of a specimen surface.

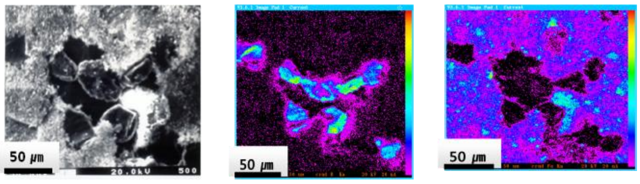


Fig. 3. Crud particles collected by an ultra-sonic system using a filter-paper.

Table 1 Quantitative analysis of boron rich face(wt %)

Ele.	Zr	Ni	Mo	O	Cr	Fe	B
Con.	4.7	3.8	3.8	3.8	0.8	1.9	Bal.

3. Conclusion

Using EPMA, composition and shape of the crud analysis was performed sampling from an irradiated nuclear fuel cladding tube. The way of crud layer measurement and distribution of specific elements on the surface of crud were discussed. Crud samples by scraped a metal knife was found that oxygen was distributed unequally, iron and nickel, which are known as the main elements of the crud, were investigated. Crud samples by cutting fuel cladding tube were observed thickness and shape. Crud samples by Ultra-sonic-system were obtained of a boron precipitate. Existence of boron and specific element should be investigated by executing an overall qualitative analysis of the surface of fuel cladding tube.

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