Crud Analytical Technique by abnormal high beam current using EPMA

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1. Introduction

Crud specimens, which were scraped from twice-burned fuel cladding in a Korean nuclear power plant, were intensively analyzed using EPMA. The principal elements of the crud were identified as Ni and Fe, where the ratio of Ni/Fe was approximately 1.3 by using EPMA. To investigate the morphology and composition of the pure metallic material composing the crud, we tried to remove coolant impurities. We tried to increase the EPMA current to an abnormal intensity until the impurities contained in the crud were melted. The technique was applied by opening an adjustable aperture device for a gun alignment adjustment. As a result, it was confirmed that the impurities contained in the crud material disappeared and only the pure metal material remained. The shape and composition of the remained crud metal elements were analyzed, and the results are shown in this paper.

2. Samples and Results

2.1 Sample preparation

A crud scraper is equipment that samples crud using suction pump and filter by scrapping the crud adhered on the fuel rod with a scraper made of Al_2O_3. The scraper of the blade type is used to scrape crud from the fuel rod, and the separated crud is transported by a Peristaltic suction pump with a capacity of 0.3hp through the hole at the bottom space, and collected in a filter with pore size of 8 ㎛.

We analyzed it in beam mode, which increases the size of the beam incident on the specimen by 5,000 to 15,000 fold. These analytical methods are suitable for cases where the slope and surface of the specimen are rough. To remove the impurities adhering to the crud, the current was abnormally raised and the analytical method of the newly produced metallic material was set to fixed mode. Note that the movement of the specimen should be stopped to accurately analyses the extremely small, round specimens. Therefore we turned off all electric motors that move the specimen stage of the EPMA. In that state, even very small specimens can be analyzed. We were able to obtain the appropriate results in this way.

2.2. Crud analysis using EPMA and SEM

Prior to carrying out this study, we conducted an EPMA analysis on several specimens. As shown in Fig. 1, the crud was shaped as W/L/t ≒ 50/120/12 ㎛, and the boiling chimney-hole size was observed to be: ≒ 6 ㎛. As for the surface shape of the area contacting the coolant, crud materials dissolved in the coolant were shown to be deposited in a precipitated form.

Precipitation growth occurs as very small particles gradually grow, rather than being deposited as a big flake. Other crud flakes are considered to be crud in areas that have contact with the cladding, as the surface is flat as it contacts the cladding, and the boiling chimney hole is not clear. As for the shape of these crud flakes, precipitations that reached a relatively consistent size were observed. In the x-ray map analyzed by the WDS of this mass, the upper part of the mass has more deposits than the lower part, and iron shows a relatively even distribution, whereas oxygen is higher in the upper part than in the lower part. As for the shape of this crud flake, precipitations that reached a relatively consistent size are observed. Whereas an x-ray map of iron and nickel analyzed by WDS of this mass shows a generally even distribution, the concentration distribution of oxygen is not shown. The results of a quantitative analysis ranging in magnification by 5,000, 8,000, and 10,000 times, are shown in Table 1. The Ni/Fe ratio of 1.4 obtained by the EPMA analysis is a typical value of the fuel crud.

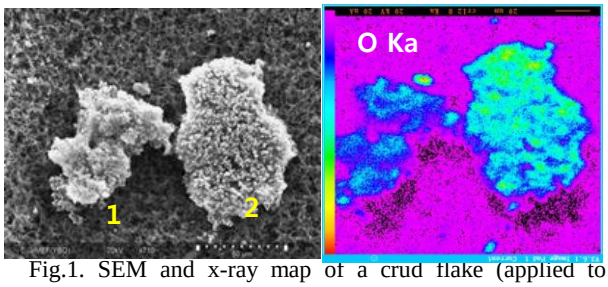

Fig.1. SEM and x-ray map of a crud flake (applied to normal beam current injection).

Table1. Chemical composition of crud flakes in area measured by EPMA analysis (at%)

No	Fe	Ni	Cr	O	Ni/Fe
No 1	18.2	24.1	2.3	54.7	1.3
No 2	39.0	57.0	0.8	2.5	1.5

2.3. Applied to abnormal high beam current injection

Although the general power application conditions for an EPMA analysis are about 20 kV and 10 nA, we increased the current conditions until the crud started to burn. The end of the power condition was 20 kV and 1200 nA. To increase the current condition, we created a current by opening an adjustable aperture device for a gun alignment adjustment on the EPMA. When the current reached 1200 nA, we could see the current value displayed on the screen. The value of the current displayed on the screen was 1200 nA; in our opinion,

however, it is assumed that the value of the current actually applied will be much higher. This is because it is difficult to generate a constant current when the adjustable aperture device for gun alignment is opened. In other words, because the current flow can be very large and variable, we estimate that the final current value applied to the crud sample was estimated to be 1,500 to 2,000 nA.

When we applied the current up to 1200 nA, we were able to identify the burning spot on the monitor. After applying the current for 5 seconds, we could observe that the shape of the crud was changed, as shown in Fig 2. We assumed that if crud burned, while burning, all non-metallic materials would be burn away. As shown in Fig. 2, it must be the material that forms most of from the crud. Therefore, metal materials of pure crud are judged to be very small in volume in the overall crud. The reason for this phenomenon is considered to be charging, which is a phenomenon that occurs due to an insufficient current flow to ground connection.

While charging, heat is created from the interaction when a large amount of electrons temporarily collides with a specimen with poor conductivity. It is considered that a part of the crud is evaporated, and the main metal composition material such as iron remains. Attention is required because the application of this condition can significant damage the device.

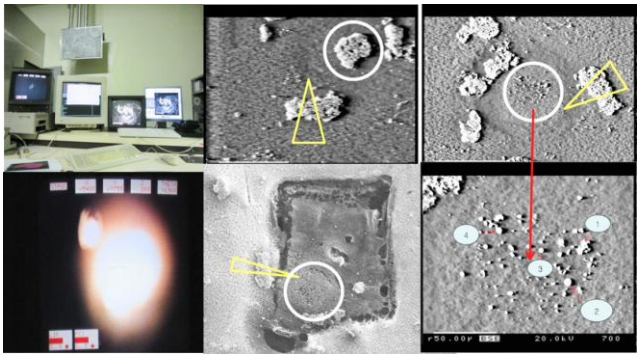

Fig. 2. EPMA monitor, burning spot on monitor, crud flake before and after burning. (20 kV, 1200 nA, beam exposure time; 5 s)

No1 shows typical nickel ferrate, which has a composition as shown in Table 2. The distributed locations of nickel, ion, and oxygen are different. In particular, iron shows a higher composition in the lower part of the specimen. Oxygen shows a similar concentration distribution as iron. According to M. Haginuma[1], the formation of nickel ferrite, which is known to be the main composition of fuel crud, is an important subject in relation to the reactor coolant chemistry, which is also formed by an exchange reaction between Ni^{2+} in the coolant and Fe^{2+} in the Fe_3O_4 lattice [1]. The results agree with M. Haginuma's paper, which we confirmed.

Table2. After burning, quantitative analyses of the remaining metals (beam conditions: 20 kV, 20 nA).

	Fe	Ni	Zn	Cr	O
No 1					
wt %	30.36	16.52	0.37	0.84	17.74
At %	27.80	14.39	0.21	0.83	56.69
No 2					
wt %	28.39	20.89	0.42	1.66	0.00
At %	56.33	39.43	0.70	3.54	**0.00**
No 3					
wt %	25.54	20.10	0.43	0.70	0.38
At %	54.20	40.58	0.78	1.61	**2.82**
No 4					
wt %	19.04	18.28	0.39	1.62	5.73
At %	32.56	29.73	0.57	2.97	34.18

3. Conclusions

We changed the power conditions for this study. Even though the general power application conditions for an EPMA analysis are about 20 kV and 20 nA, the power conditions applied to this analysis were 20 kV and 1200 nA during 5 to 30 seconds As a result, we found that part of the crud was evaporated, and the main metal composition, such as iron, remains. The ratio of pure metal, compared to the volume of impurities and other water-soluble amorphous materials, is absolutely low in the entire volume of the crud. Fuel crud is generally composed of $NiFe_2O_4$, NiO, Ni^0 etc[2]. We tried to quantitative analyses the remaining metallic crud precipitate No1~4. We found $NiFe_2O_4$ at No1 specimen, and boll type pure Ni/Fe material at No3, also we can see Fe/Ni/O = 1/1/1 at No4 specimen. The results from this experiment may be different from the composition of crud. However, we believe that this method presents a new way of analyzing crud.

REFERENCES

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