Comparison of quantitative analysis methods on CRUD using an EPMA and X-ray image mapping

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1. Introduction

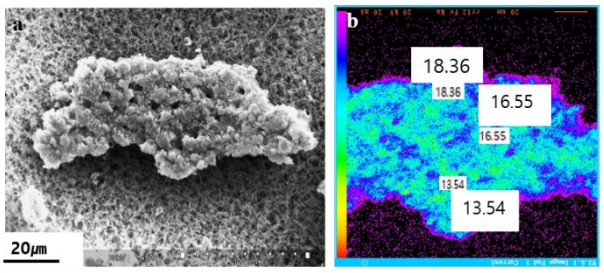
CRUD specimens were analyzed using an electron probe micro analyzer (EPMA) and X-ray image mapping. It is difficult to analyze the composition of radioactive corrosion products using an EPMA due to the size and rough shape of surfaces. It is particularly challenging to analyze the composition of radioactive corrosion products in the form of piled up, small grains. However, useful results can be derived by applying a semi-quantitative analysis method using an EPMA with X-ray images. A standard-less, semi-quantitative method for wavelength dispersive spectrometry (WDS) EPMA analysis was developed with a view to simplifying the analytical procedure required.

In this study, we verify the reasonable theory of semi-quantitative analysis and observe the semi-quantitative results using a good surface condition sample. Based on the validated results, we analyze the highly rough-surface radioactive corrosion products and assess their composition. Finally, the usefulness of the semi-quantitative analysis is reviewed by verifying the analysis results of radioactive corrosion products collected from spent nuclear fuel rods.

2. Results and discussion

Electron probe micro analyzers (EPMAs) are widely applied in the analysis of chemical compositions of unknown materials, especially for irradiated nuclear fuels [1]. Quantitative analysis in EPMA analysis is usually based on a comparison of the intensities of a characteristic line emitted from a sample and from a standard of known composition. This process consists of first selecting specific lines of the elements, with the hypothesis that these elements are present in the sample, and second, measuring the X-ray intensities emitted from the unknown and the standard samples. This second step requires much time and sometimes poses problems if proper standards are not available [2,3].

Semi-Quantitative Analysis. A standard-less, semi-quantitative method for EPMA using wavelength dispersive spectrometry (WDS) was developed with a view to simplifying the analytical procedure required with this methods [4]. Based on spectrum acquisition, this method provides a way to obtain the sample composition in a short time with the advantages of the WDS system and with reasonable accuracy.



Item	Fe	Ni	Cr	Zn	O	Total
1	14.62	28.63	2.15	0.93	12.51	58.84
2	17.60	22.73	1.83	0.72	10.24	53.13
3	15.95	19.63	2.04	0.65	9.49	47.76
Ave	16.06	23.66	2.01	0.77	10.75	53.24

Fig. 1 SEM and X-ray image mapping of Fe in the radioactive CRUD flake specimen

The CRUD specimens shown in Fig. 1 are several tens of micrometer in size, and the roughness of the surface is also very rough, as shown in the figure [4]. The data analyzed by EPMA analysis using a standard specimen are shown at the bottom of the figure. Due to the surface roughness of the sample, the overall average composition for the three points was about 53 wt%.

This analysis method is not appropriate because it is a method of analyzing after checking the surface condition of a sample with SEM images. However, as shown in Fig. 1(b), it is determined to be a usable analysis method because the semi-quantitative result values for the desired site are checked with the identification of specific parts and shapes of samples identified by X-ray image mapping. Rather than analyzing the composition while observing the surface of the sample with SEM, as shown in Fig. 1(b), there is the convenience of checking the composition by simply clicking the desired point while viewing the X-ray image mapping. Of course, it was confirmed that the semi-quantitative analysis result has an acceptable value. In Fig. 1, the composition of Fe analyzed using the standard specimen was 16.06 wt%. In addition, it was confirmed that the composition of Fe

analyzed by the semi-quantitative method had a useful value of 16.15 wt%.

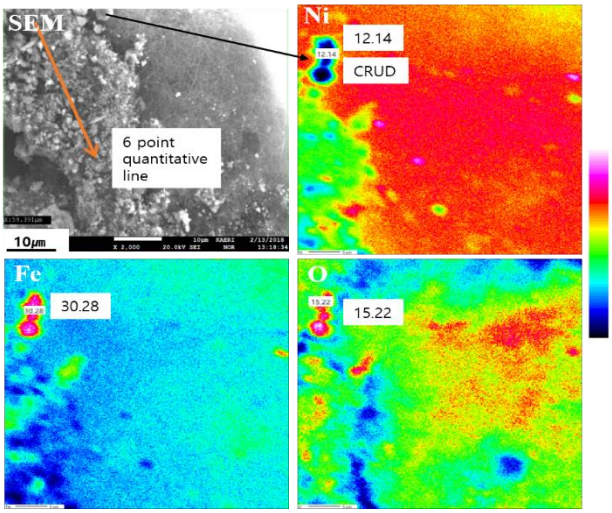


Fig. 2. SEM of a CRUD flake sampled with ultrasonic cleaner in a pressurized water reactor plant, and X-ray image mapping of Ni, Fe, O

Fig. 2 shows the SEM shape of typical corrosive products bonded to nuclear fuel cladding and the result of Ni, Fe, and O X-ray image mapping. In the figure, among the corrosive products of various forms of debris, the existence of CRUD, which nuclear power plant workers and researchers are interested in, was identified. As shown in Fig. 2. The analysis results in 9.12 wt% Ni, 8.62 wt% Fe, and 8.98 wt% O. The results of this were not identified as the chemical composition of the CRUD, NiFe2O3.x (18.65 wt% Ni, 32.45 wt% Fe, and 17.68 wt% O).

3. Conclusions

Highly rough-surface radioactive corrosion products and reviewed their composition. The usefulness of semi-quantitative analysis can be reviewed by verifying the analysis results of radioactive corrosion products collected from spent nuclear fuel rods. In fact, when nuclear power plants have the need to confirm the composition of a material having such an irregular shape, when analyzing the composition of a sample using an EPMA, it should be recognized that the only method of analysis is a semi-quantitative method. Again, the semi-quantitative method is a useful method for radioactive corrosion products with severe surface roughness.

References

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