Analysis of a NiO Precipitates in the Crud Using EPMA

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1. Introduction

Crud (Chalk River Unidentified Deposit) radioactive corrosion products formed inside nuclear reactors is a major problem in commercial power-producing nuclear reactors. It can be transported onto the reactor core by cooling water, where they can be deposited on the outside of the fuel pins to form crud. If these radioactive deposited break loose and circulated through the plant, they can cause safety hazards for plant workers [1][2]. Crud has been taken from the spent fuel cladding and analyzed by an EPMA (Electron Probe Micro Analyzer).

Boron was found in the crud samples taken from the fuel cladding and assemblies. In this study, Boron has been analyzed by EPMA techniques to gather Boron information related to composition, structure, and morphology.

2. Samples and Results

2.1 Samples

The crud samples in this study are taken from the U.J. NPP reactor by an ultra-sonic system and the Y.G steam generator tube plug,whese samples were cracked by PW-SCC. EPMA crud samples were prepared by transferring a few particles of crud to a carbon sticky dot on a standard sample holder using tweezers and a small paintbrush in a glove box. Individual particles were too small to readily see and manipulate, and these was no charging effort without coating.

2.2. EPMA

The EPMA(Electron Probe Micro-Analyzer, SX-50R, CAMECA, France) used in this experiment can treat irradiated nuclear fuel by a special shielding of the specimen holder and is specifically shielded with lead and tungsten to permit the analysis of irradiated nuclear fuel. The maximum radiation activity in this EPMA is allowed to be below to 3.7×10^{10}Bq. The condition of EPMA was 20 kV of an electron acceleration potential and 20 nA of a beam current.

2.3 Results

Fig. 1 shows crud layers on the cladding surface. The crud layer which shows bleak dots that form on the surface, as shown in Fig.1, is often referred to as a duplex oxide film containing a very stable inner- chromium-rich oxide, although recent researchers suggest the outer layer also has a Cr component.

Fig.2and3. show SEM of crud deposits from the cladding interface and steam generator tube plug respectively. Compare the results of the two SEM image, we can see there are similar same surface like the needles figures. For this reason, the soluble nickel ions which primary coolant water in the reactor are combined with oxygen form flow not only on the cladding but also steam generator tube plug is accrued.

Fig. 1. Crud layers on the cladding surface

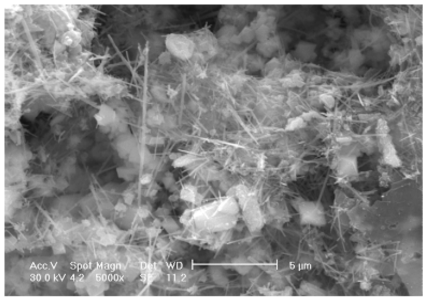

Fig.2. SEM of crud deposits from the cladding interface

Nickel ferrite was the most common component of the crud. The amount of nickel substitution was high in crud from all of the plants, having a composition close to the mineral Trevorite, NiFe2O4. The nickel ferrite crystals were typically blocky and displayed well-defined crystal faces [3].

A needle shapes shown in Fig.3 which is dissolved in the reactor coolant 1st solution in the shape of a nickel oxide. We can found of the shapes of nickel needle on Fig.3 and Fig 4 shows the same, which crud deposits shapes from reactor's cladding crud and steam generator tube plug shape .

Fig.4 shows image mapping of the surface on Fig.2 for oxygen. In this figure we can see the entire distribution of the oxygen on the sample surface. Fig.4 is the results of image mapping adjacent area in Fig.2. But Fig. 4 is not represented in the form of shaped like a needle because the needle is in the form of hexagonal shape of the cone. This can be explained by the principles of analysis equipment.

When the scattered electron beam incidents to the needle shape, reflected x-ray on the surface of the crud does not recognized equally for the needle shaped in NiO. In Figure 4, oxygen was distributed evenly of the overall as an oxides form. Table 1 is a quantitative analysis result of the adjacent Fig.2 areas by the EPMA analysis of the surface applied Tv Mode (x 500) techniques analyzed. Tv Mode analysis techniques is applied to the rough surface samples and the incident beam reflected on the sample surface to wide shot, we can get the approximate composition of the sample, but the exact composition is hard to obtain.

As shown in Table 1 the entire content of the sample is around 60 wt%, this quantities results of the sample can determine the approximate composition of the crud deposits from cladding interface.

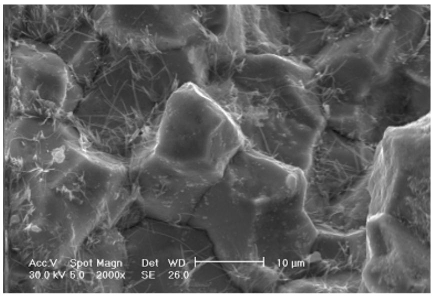

Fig.3. SEM of crud deposits from the steam generator tube plug

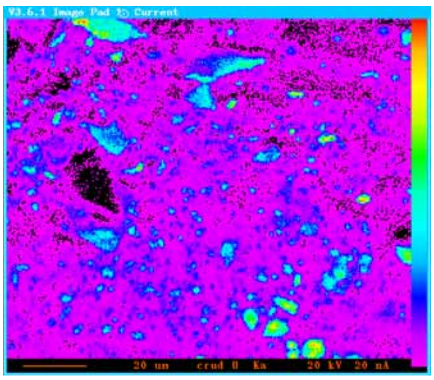

Fig.4. Image Mapping of crud deposits from the cladding interface

Table 1. Concentrations of crud taken from cladding interface (unit: wt.%).

Elements	Ni	Fe	Cr	Zr	O	Si
Wt %	45.5	2.3	0.8	1.1	10.4	0.3

However, needle-like nickel oxide particles were sometimes found in association with the more typical blocky NiO particles.

High temperature sampling of reactor coolant water determined that metallic nickel is a common component of the circulating crud. The dissolution of such metallic nickel particles is no doubt responsible for a large part of nickel released during a typical shutdown.

NiO was absent from the high temperature sampling, but present in several of the scraped crud samples. This suggests that the NiO content of RCS surfaces was low, and the NiO that does exist may be concentrated on the core.

3. Conclusion

Composition of crud taken at the spent fuel rod discharged from Y.G NPP was determined by EPMA. High sub-cooled boiling rates increased crud deposition, but did not completely explain the development of thick crud.

Other factors, such as the quantity of corrosion products transported to the core and the distribution of boiling within the core were no doubt involved.

A number of new crud features were discovered in this work. These included layering with the crud as well as the presence of needle-like crystals.

References

[1] R. RIESS, "Chemistry Experience in the Primary Heat Transport Circuits of Kraftwerk Union Pressurized Water Reactors" Nuclear Technology 29, 153-159 (1976)

[2] PWR Axial Offset Anomaly (AOA) Guidelines: Revision 1, EPRI, Palo Alto, CA: 2004. 1003213

[3] Root Cause Investigation of Axial Power Offset Anomaly, EPRI, Palo Alto, CA: 1977. TR-108320