Study of the Composition of Cruds on the High Burnup Fuel Cladding in PWR

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1. Introduction

Long-term and high burnup fuel cycles cause deposits on the cladding surface by the solubility change of the coolant water, which depends on both pH and temperature [1]. Crud formation occurs both through direct deposition on the suspended solid particles in the coolant water, and through 'evaporate and dry-out' mechanism. The latter is named 'Marangoni Flow' which can affect the solute concentration distribution in the bubble interface near the heating wall [2].

According to results of the post irradiation examination on PWR rods in KAERI, the thickness of the crud layer was shown to be maximum 15 μm in 55 GWd/tU rod average burnup. This paper describes results of a quantitative determination of the crud composition measured by EPMA. The samples were taken from the high burnup fuel rods which were irradiated in a Korean commercial PWR.

2. Methods and Results

The quantitative analysis on the crud composition was performed by EPMA with radiation shielding walls which protect the human body irradiation in KAERI.

2.1 Sample preparation

Cladding and fuel materials of the sample were zircaloy-4 alloy and UO_2. The average fuel rod burnup was 40.5 GWd/tU and the average pellet burnup was 47.8 GWd/tU at the sampling position, the height 2980mm from the rod bottom. Prior to mounting the sample, removing a pellet inner cladding was performed by using the drilling machine in order to minimize radioactivity of the sample.

2.2 EPMA

Electron Probe Micro Analyzer (EPMA), CAMECA-50R has been used to analyze crud on cross-sectional ring sample of the fuel cladding. This make possible to measure a weight percent from Be to U included in samples with two Wavelength Dispersive Spectroscopes (WDS) and four crystals.

2.3 Crud composition

Point and line scan on the crud outside cladding were done by EPMA. Figure 1 shows granular agglomerates of crud deposited on the cladding surface including analysis points and the position of line scan. The crud layer is approximately 10 μm. The crud consists of crystals with a mean size of about 1 μm and sharp edges.

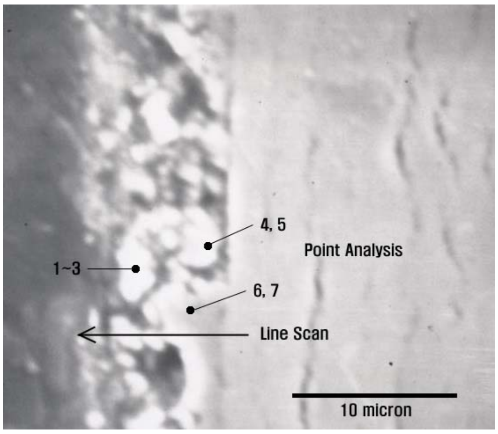

Figure 1. Granular agglomerates of crud deposited on the cladding surface

The chemical composition data by the point analysis are given in Table 1. The chemical composition of point 6 accords with nickel ferrite ($NiFe_2O_4$) as reported in some papers as a main crystal of crud [3]. At agglomerates of the outer crud, point from 1 to 3, they are likely to consist mainly of hematite (Fe_2O_3) except nickel. Nickel to iron of most measuring points is approximately shown to be 0.5. Some metallic elements; Cr, Mn, Co, Cu, Zn are relatively minor.

Table 1. Chemical compositions by the point analysis

at%	O	Fe	Ni	Cr	Mn
#1	47.407	33.468	17.418	0.836	0.693
#2	47.337	33.843	16.939	1.086	0.602
#3	45.708	34.901	17.702	0.664	0.762
#4	81.586	11.415	6.154	0.11	0.31
#5	69.633	18.837	10.563	0.141	0.586
#6	56.3	27.671	15.181	0.579	0.162
#7	62.329	23.077	13.635	0.385	0.251

In order to make an analysis on the composition distribution of crud elements along the depth of a crud layer, the line scan on crud was tried. Figure 2 shows results of the line scan. Nickel ferrite ($NiFe_2O_4$) is regarded as a major crystal in the inner part of the crud, close to the oxide layer whereas in the outer crud hematite (Fe_2O_3) presents generally.

Nickel to iron atomic ratio has tendency to increase as a measuring point moves from the inner part to the outer part of the crud layer. Nickel to iron ratio has been reported to increases as the temperature rising [4]. This hints operating conditions of a nuclear fuel. Also according to Ni-58(n,p)Co-58 reaction, Ni-58 (naturally, 68%) is the parent element of Co-58, which is a major radiation source with a half life of 70.86 days. This could increase the occupational radiation exposure during the operational period at nuclear power plant.

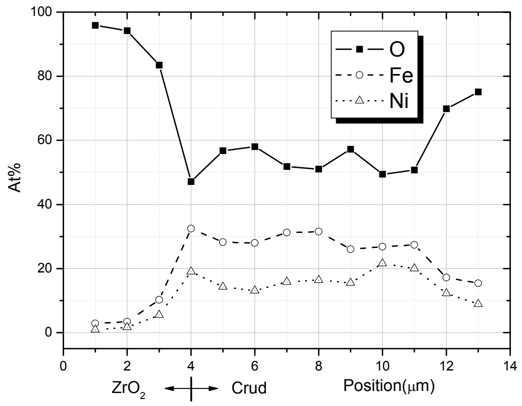

Figure 2. Chemical composition distribution by the line scan

3. Conclusion

Nickel ferrite ($NiFe_2O_4$) is seemed to be main chemical form of PWR crud. Crud compositions in the inner part of the crud layer mostly accord with nickel ferrite. Potential radioactive element, nickel increases as a measuring point moves to the outer part of the crud layer. This can hint operating conditions of a nuclear fuel.

Crud formation on the fuel cladding surface become more serious problem as the discharge burnup of the nuclear fuel increases due to an economic advantage. Therefore more extensive examinations are required to search the mechanism of the crud formation and to protect the crud deposition during the operation period.

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