



FR0301277



\*DE019468881\*



FR0301277

NIS-FR-1665

## Experiences of Ultra-Low-Crud High-Nickel Control in Onagawa Nuclear Power Station

M. Saito, Y. Goto, T. Shinomiya, M. Sato Tohoku Electric Power Company, Inc.  
K. Yamazaki, H. Hirasawa, T. Yotsuyanagi Toshiba Corporation

### 1. Introduction

We have adopted various countermeasures for worker dose reduction to plants in Onagawa Nuclear Power Station. "Ni/Fe ratio control" has been adopted to Unit 1, and "ultra-low-crud high-nickel control" has been adopted to Unit 2 and 3, along with other countermeasures like wide utilization of low Co materials, for the purpose of dose rate reduction of primary recirculation piping which is thought to be one of the main exposure sources.

In this paper, we describe, first, the reason and background that ultra-low-crud high-nickel control has been adopted to Unit 2, and, second, water chemistry of Unit 2 up to the 5th cycle under ultra-low-crud high-nickel control compared to that of Unit 1 under Ni/Fe ratio control. Following those, we show brief analysis of the fuel crud of Unit 2 and water chemistry of Unit 3 only at the startup stage.

### 2. Adopting ultra-low-crud high-nickel control to Onagawa Unit 2

In the history of Onagawa Nuclear Power Station, the Ni/Fe ratio control has been adopted to Unit 1, 524MWe BWR-4, almost from the middle of its first cycle.

The Ni/Fe ratio control is a technology for dose rate reduction of piping through concentration reduction of activated Co in reactor water. The control is performed by controlling feed water Fe crud, and Co concentration in reactor water is reduced by locking Co up on the fuel surfaces on which Co is incorporated in stable ferrite oxides. Unit 1 has precoating-type condensate water purification system, and ratio control is attained through setting the ratio of Ni ion and Fe crud in feedwater by changing the density of precoating. This technology was quite effective in dose rate reduction, and has been adopted to many Japanese BWR plants by the same methods as Unit 1 or bypassing condensate water filters. (Ref. 1)

However, before Unit 2, 825MWe BWR-5, started its operation, Co-60 concentration in reactor water and dose rate on piping of brand-new Japanese BWRs increased in spite of the Ni/Fe ratio control. (Ref. 2) This phenomenon occurred after zirconium liner fuel (so called Step 1 type fuel) came to BWRs, and was caused by chromium enrichment on mechanically polished and non-autoclaved surface of zirconium liner fuel rods. The chromium enhances Co-60 dissolution from fuel crud by lowering local pH on the surfaces. (Ref. 3)

Unit 2 also started its operation with zirconium liner fuels for its initial core; therefore, an alternative to the Ni/Fe ratio control was requested for its water chemistry control. Water chemistry of Unit 1 had been operated for more than ten years with rather low dose rate of primary recirculation piping at that time. We evaluated Unit 1 chemistry, and our conclusion of the evaluation was that neutral pH of reactor water and existence of small amount of zinc in reactor water, released from condenser tubing, was the main reason of the good performance of Unit 1. The natural zinc addition is not expected in Unit 2 because Unit 2 has titanium condenser tubing. The result of more detailed analysis of Japanese Ni/Fe ratio control plants indicated that the existence of Ni ions in reactor water suppressed Co deposition onto primary piping surfaces. That was confirmed by laboratory test, which deposition on stainless surfaces was suppressed by Ni ions. (Ref. 1)

The anxiety of increase of activated Co concentration in primary coolant came up unless the Ni/Fe ratio control was adopted. The initial core of Unit 2 consisted of Step 2 type fuels. They are also zirconium liner fuels; however, their spacer springs are decreased in surface areas to 1/3 of Step 1 type fuels, are prefilmed by a high temperature air oxidation process and contain lower Co. The use of Step 2 type fuels was not expected to induce extreme increase of Co concentration in reactor water. (Ref. 4)

Unit 2 has a full-flow hollow fiber filter system in the condensate water purification system, which can easily control crud concentration. As a result, we decided the ultra-low-crud high-nickel control be adopted to Unit 2 from the beginning of startup operation. The high Ni ion concentration has been obtained by controlling crud (Fe) concentration extremely low because excess Ni in terms of ferrite formation remains in reactor water. By the ultra-low-crud high-nickel control, not only dose rates of high temperature piping on which ionic cobalt is deposited but also total worker dose has been expected to be reduced. (Ref. 1, 4)

Based on the successful experiences of the ultra-low-crud high-nickel control in Unit 2, we have started adopting this technology to Unit 3 that are starting its commercial operation in January 2002.

### 3. Water chemistry and dose rate behaviors in Onagawa Unit 2

#### 3.1 Behaviors of Fe and Ni

Unit 2 has a full-flow hollow fiber filter for condensate purification system. Feedwater crud concentration can, therefore, be controlled at ultra low level by combination with feedwater oxygen injection to suppress corrosion of piping and heater trains.

Fig. 1 shows insoluble Fe concentration in feedwater of Unit 2 by the 5th cycle with that of Unit 1. The concentration of Unit 2 has been controlled well below 0.1 ppb, which is much lower than that of Unit 1 under the Ni/Fe ratio control, 0.1–1.0 ppb.

Soluble Ni concentration in reactor water has been maintained from 2 ppb to 4 ppb as shown in Fig. 2, which is much higher than that of Unit 1. After the 4th cycle, the concentration has jumped up around 4 ppb. This jump-up corresponds to the decrease of insoluble Fe in feedwater. The decrease of Fe in feedwater results in the decrease of consuming Ni in reactor water.

At the plant where the Ni/Fe ratio control is adopted, Ni ion concentration in reactor water is maintained around 0.2 ppb that corresponds to the solubility of Ni ferrite under BWR condition. On the other hand, soluble Ni concentration in reactor water of the ultra-low-crud high-nickel control plant is maintained at a higher level because of the existence of excess Ni in terms of Ni ferrite stoichiometry.

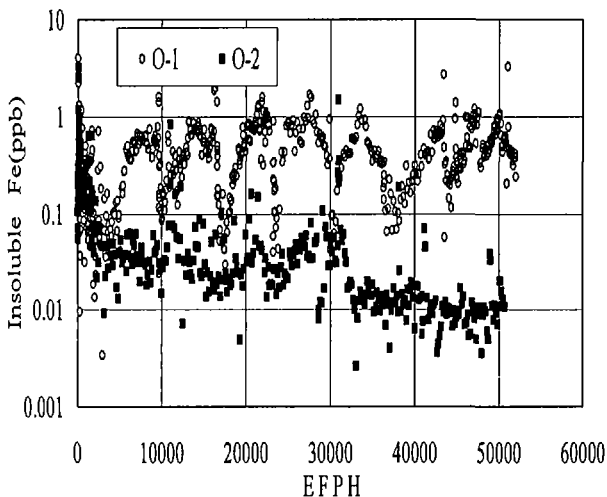


Fig. 1 Feedwater insoluble Fe concentration of Onagawa Unit-1 and 2 : ultra low crud control was adopted at Unit-2

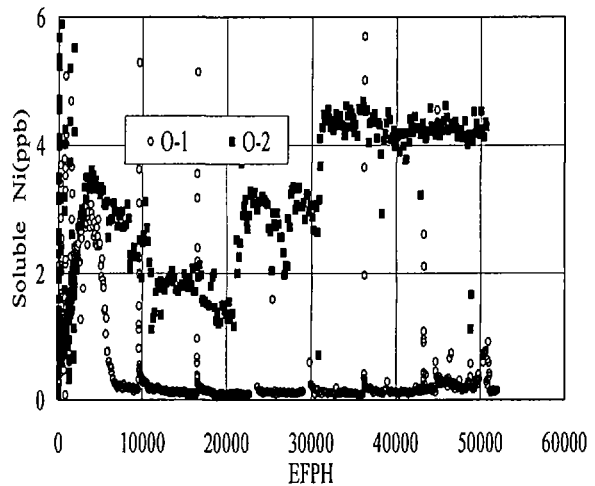


Fig. 2 Reactor water soluble Ni concentration of Onagawa Unit-1 and 2 : Ni concentration was kept high by ultra low crud control

### 3.2 Behaviors of activated Co and Mn

Fig. 3 shows soluble Co-58 concentration in reactor water of Unit 1 and Unit 2.

Fig. 4 shows that of Co-60. The activated Co concentration of Unit 2 under the ultra-low-crud high-nickel control is generally higher than that of Unit 1 under the Ni/Fe ratio control. The concentrations tend to saturate after the 3rd cycle, and are expected to be around 20-40 Bq/ml for Co-58 and 6-8 Bq/ml for Co-60.

Fig. 5 shows soluble Mn-54 concentration in reactor water of Unit 1 and Unit 2. The ultra-low-crud high-nickel control prevents Fe crud activation on fuel surfaces and Mn-54 concentration in reactor water decreases.

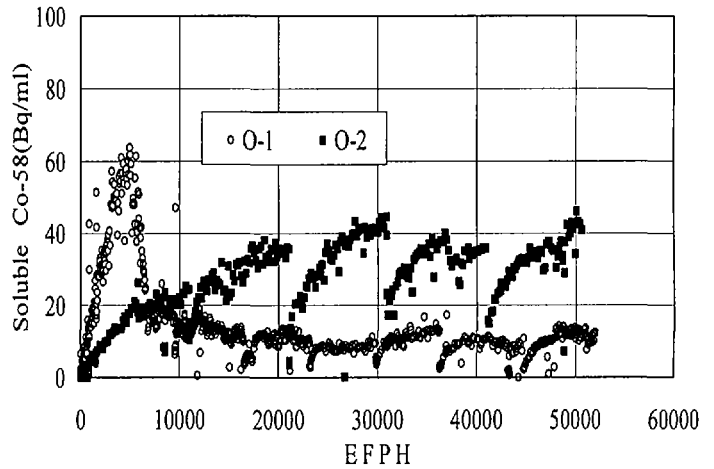


Fig. 3 Reactor water soluble Co-58 concentration of Ongawa Unit-1 and 2 : Co-58 at Unit-2 is relatively high because of ultra low crud control

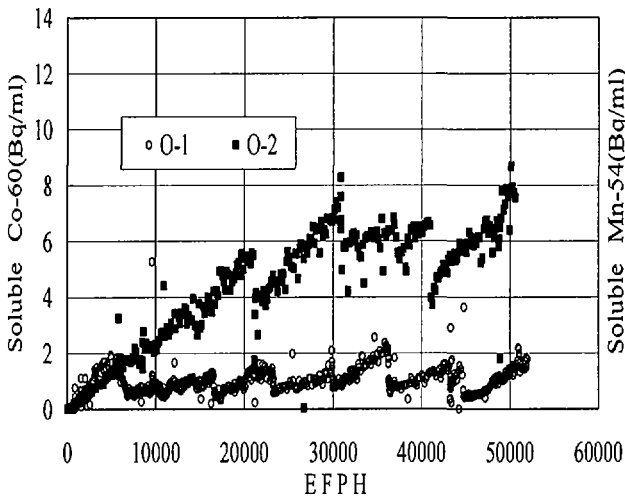


Fig. 4 Reactor water soluble Co-60 concentration of Ongawa Unit-1 and 2 : Co-60 at Unit-2 is relatively high because of ultra low crud control

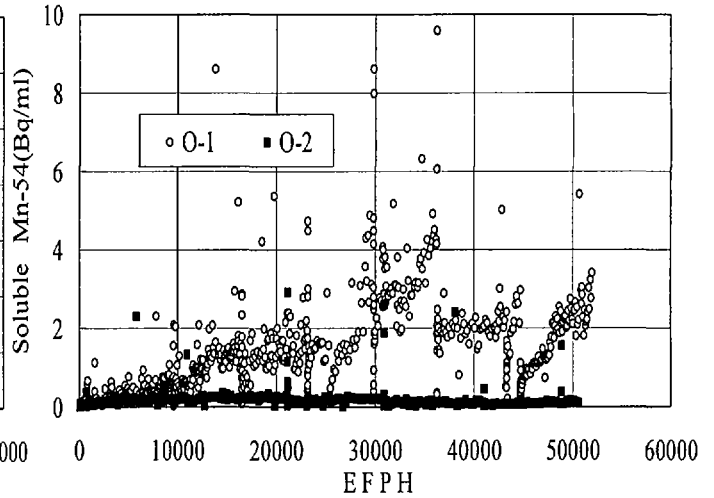


Fig. 5 Reactor water soluble Mn-54 concentration of Ongawa Unit-1 and 2 : Mn-54 at Unit-2 is relatively low because of ultra low crud control

### 3.3 Trends of dose rate at primary recirculation piping

The dose rate of primary recirculation piping of Unit 2 is compared to that of Unit 1 as shown in Fig. 6. The dose rate of primary recirculation piping of Unit 2 was extremely low at the first outage. It was getting slightly high up to the 5th outage and resulted in 0.56 mSv/h that is higher than that of Unit 1. It is still lower than those of typical Japanese BWR plants although it should be watched for a while.

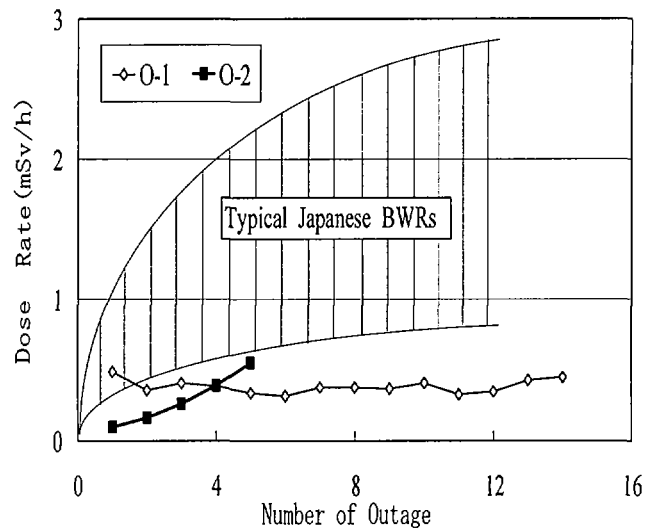


Fig. 6 Dose rate of primary recirculation piping of Ongawa Unit-1 and 2

## 4. Features of activated Co behaviors under ultra-low-crud high-nickel control

### 4.1 Deposition rates onto piping

Under the ultra-low-crud high-nickel control, deposition rate of Co-60 onto primary system piping should be lowered due to the effect of soluble Ni in reactor water. The deposition rate of Co-60 can be calculated by dividing deposited amount of Co-60 on piping by average concentration of Co-60 in coolant during operational time.

The results for primary recirculation piping of Unit 1 and Unit 2 are shown in Fig. 7. The deposition rate of Co-60 onto the primary recirculation piping in unit 2 has been kept quite low.

### 4.2 Deposits on fuel cladding

The amount and characteristics of deposits on fuel rod surfaces are key factors in terms of better understanding of Co-60 behavior under the ultra-low-crud high-nickel control. For a fuel rod removed from the core of Unit 2 at the 4th annual outage, deposits were sampled and analyzed to determine the amount of transition metals such as Fe, Ni, and Co. The procedure of sampling was the same as described in Ref. 4; the deposits were scraped first by a plastic blade for soft crud and then by an alumina stone for hard crud, and the two types of crud were totaled.

The results are summarized in Fig. 8. All the fuel rods are made of the BJ type Zircaloy tube. Z1E235, Z1F601 and Z1F228 represent the data of the rods removed from Unit 1 under the Ni/Fe ratio control, and Z2X034 and Z2X035 represent those from Unit 2 under the ultra-low-crud high-nickel control. Z2X034 was removed at the first outage of Unit 2, and Z2X035 was at the 4th outage. Except for Z2X035, they were reported in Ref. 4. For Z2X035, the amount of Ni and that of Fe are almost same. This indicates excess existence of Ni in terms of Nickel ferrite. Because of the extremely low Fe input from the feedwater in Unit 2, the amounts of Fe for Z2X034 and Z2X035 are less than 1/10 of those for rods installed in Unit 1.

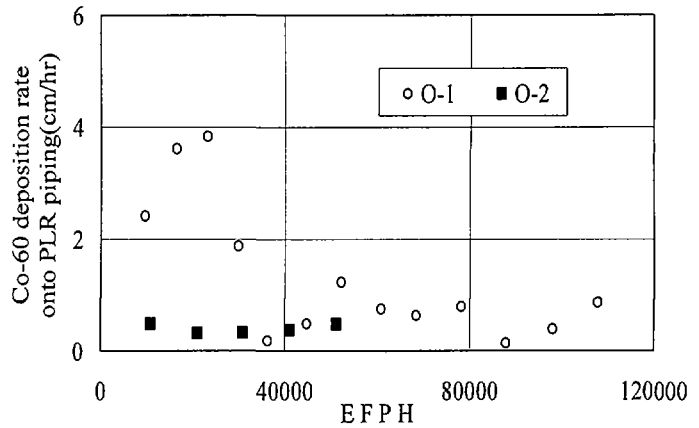


Fig. 7 Deposition rate of Co-60 onto the primary recirculation piping of Onagawa unit-1 and unit-2 (O-1: Ni/Fe ratio control, O-2: ultra low crud control)

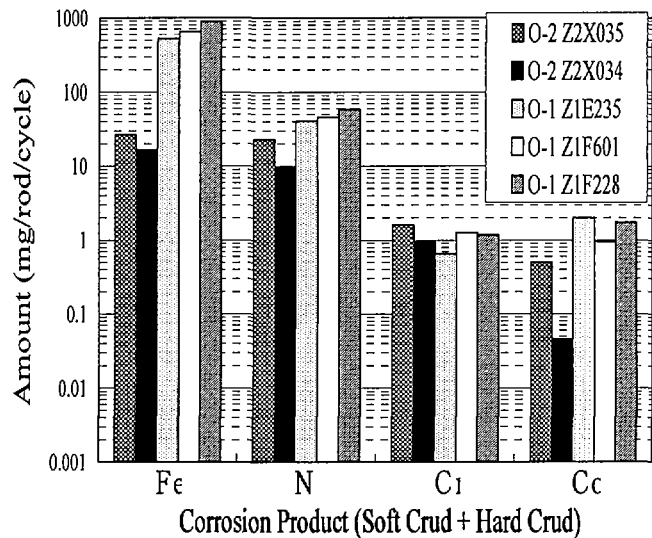


Fig. 8 Amount of deposited transition metal on fuel rods installed in Onagawa unit-1 and unit-2 (O-1: Ni/Fe ratio control, O-2: ultra low crud control)

### 4.3 Characteristics of Co incorporated in Oxides

Based on the data obtained in Unit 2, it is speculated that excess Ni plays significant roles in terms of Co-60 deposition onto piping and Co-60 behavior on fuel rod surfaces.

On primary recirculation piping, excess Ni in reactor water should contribute the formation of stable Ni ferrite layer. Due to the stability of this layer, Co-60 incorporation into the inner layer will be suppressed and then lower deposition rate of Co-60 is obtained in Unit 2.

On fuel rods, the amount of deposited Fe is quite small. This leads to the existence of Ni in reactor water and then Ni can be precipitated as NiO in boiling region. Due to rather high solubility of NiO under BWR reactor water condition, it is speculated that specific activities of Ni and Co incorporated in NiO are lower than those in Ni ferrite. As shown in Fig. 9, specific activities of Co-60 and Co-58 for Z2X035 are lower than those for other fuel rods of Unit 1.

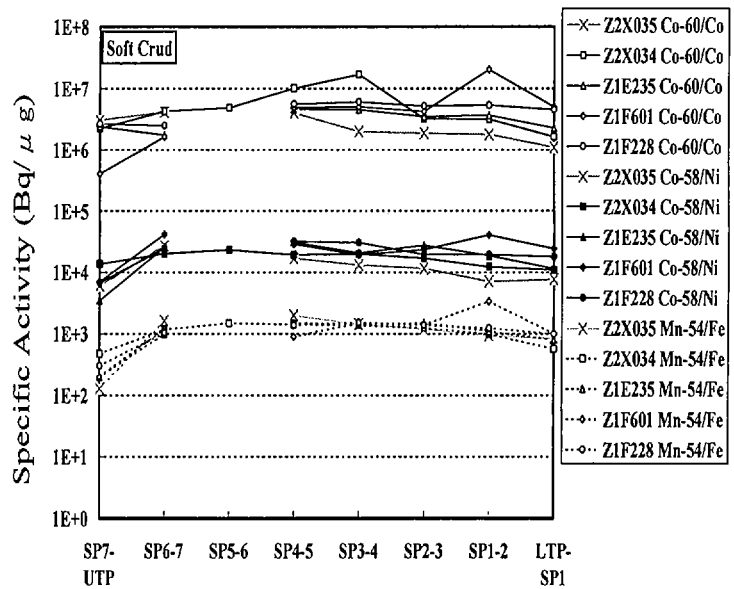


Fig. 9 Specific activities of activated transition metal incorporated in the loosely deposited crud on fuel rods installed in Onagawa unit-1 and unit-2

In terms of mechanism of Co-60 behavior under the ultra-low-crud high-nickel condition, information on stabilities of Ni compounds such as Ni ferrite and NiO is relatively important. However, such data are not enough to evaluate the mechanism of Co-60 under the ultra-low-crud high-nickel condition qualitatively. Further efforts to accumulate the data of oxides are necessary for better understanding of the role of Ni under ultra-low-crud high-nickel condition.

## 5. Water chemistry and dose rate behaviors in Onagawa Unit 3

### 5.1 Behaviors of Fe and Ni

Fig. 10 shows insoluble Fe concentration in feedwater of Unit 2 until 4000 EFPH and Unit 3 until 2000 EFPH, and Fig. 11 shows soluble Ni concentration in reactor water in the same period. In the period, crud input through feedwater is suppressed in every plant. The trends of Unit 3 are similar to those of Unit 2.

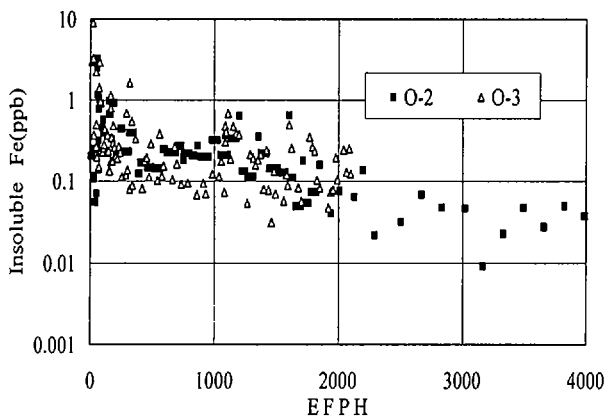


Fig. 10 Feedwater insoluble Fe concentration of Onagawa Unit-2 and 3 by 2000 EFPH

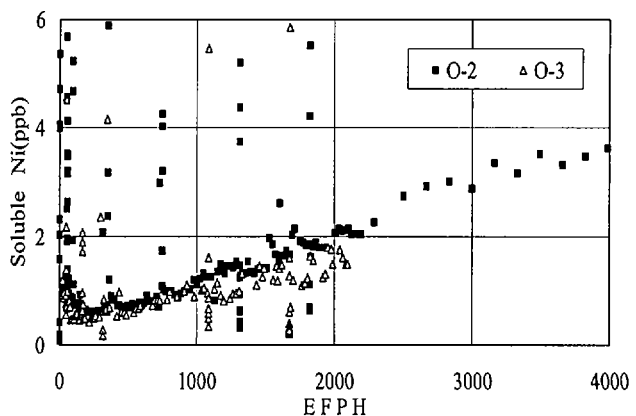


Fig. 11 Reactor water soluble Ni concentration of Onagawa Unit2 and 3 by 2000 EFPH

## 5.2 Behaviors of activated Co

Fig. 12 shows soluble Co-58 concentration in reactor water of Unit 2 until 4000EFPH and that of Unit 3 until 2000EFPH. Fig. 13 shows that of Co-60. The Co-58 concentration of Unit 3 is at the same level to that of Unit 2, and the Co-60 concentration of Unit 3 is slightly lower than that of Unit 2. This shows that Unit 3 is almost equivalent to Unit 2 in terms of the ultra-low-crud high-nickel control. The reasons why the concentrations of activated Co in Unit 2 and 3 are lower than that of Unit 1 are resulted from the improvement of reactor structural materials, enhanced utilization of low Co materials and the increase of the capacity of reactor coolant clean up system.

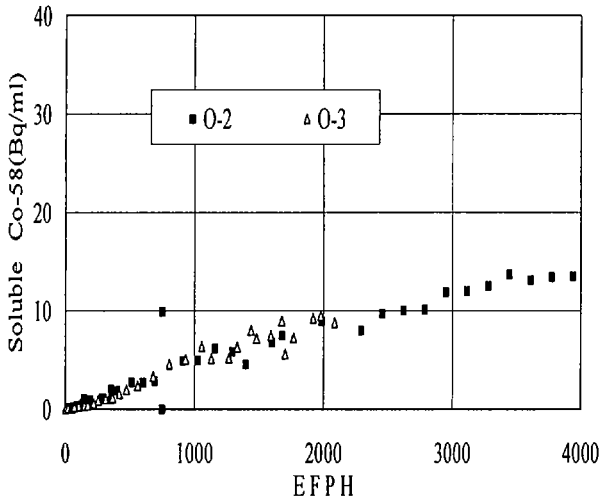


Fig. 12 Reactor water soluble Co-58 concentration of Ongawa Unit2 and 3 : Co-58 at Unit-2 and 3 is relatively high because of ultra low crud control

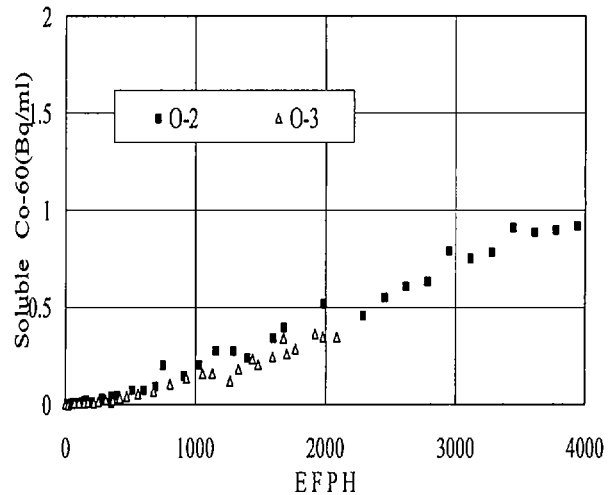


Fig. 13 Reactor water soluble Co-60 concentration of Ongawa Unit2 and 3 : Co-60 at Unit-2 and 3 is relatively high because of ultra low crud control

## 5.3 Trends of dose rate at primary recirculation piping

The dose rate of primary recirculation piping of Unit 3 is compared to that of Unit 2 as shown in Fig. 14. Until 1700 EFP H, the dose rate of Unit 3 is almost same, but slightly lower than that of Unit 2. The dose rate of Unit 3 is expected to follow that of Unit 2 since the design parameters are same.

## 6. Summary

The ultra-low-crud high-nickel control has been adopted to Onagawa Unit 2 and Unit 3 from the beginning of their start-up operations.

In Unit 2, the dose rate at the primary recirculation piping has been kept low due to the low deposition rate of Co-60 onto the piping. Due to the low input of Fe from the feedwater under 0.1 ppb as ultra low crud, Ni concentration in the reactor water has been kept around a few ppb as high nickel. High Ni condition should cause the low deposition rate of Co-60 onto piping.

Under high Ni condition, Ni can precipitate on fuel surfaces as NiO. The analysis of deposits on fuel rods indicates the existence of NiO. However, due to the instability of NiO, Co-60 and Co-58 in NiO can be easily released into reactor water. Concentrations of Co-60 and Co-58 in the reactor water have been high; however, the trend of the concentrations shows a plateau and is expected to be in the same level for future.

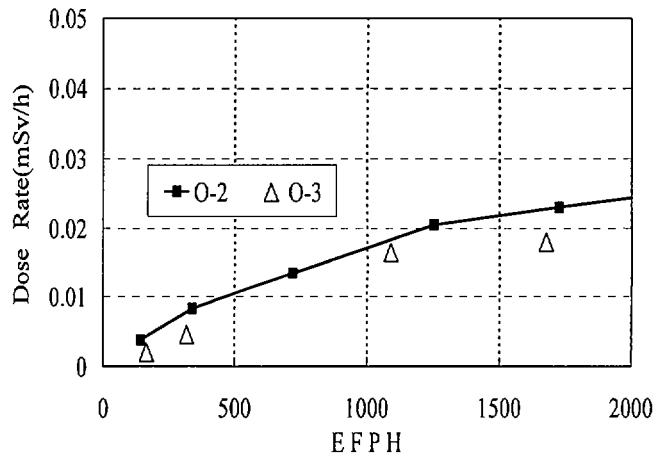


Fig. 14 Dose rate of primary recirculation piping of Onagawa Unit-2 and

As a result, the dose rate of the primary recirculation piping of Unit 2 is expected to be kept low.

In Unit 3, the ultra-low-crud high-nickel control has just started. From the data obtained during the start-up period of Unit 3, it is expected that Unit 3 also shows lower level of dose rate at the primary recirculation piping.

### **Acknowledgement**

The authors would like to acknowledge the invaluable suggestion and fruitful discussion of Emeritus Prof. Ishigure of University of Tokyo.

### **Reference**

1. K. Kawamura, S. Abe, K. Honda, K. Gotoh, T. Yotsuyanagi, K. Yamazaki and Y. Morikawa, "The history of water chemistry of Onagawa Unit-1 and Unit-2", Proc. Water Chem. Nucl. React. Sys. 96, p182, 1996
2. K. Ishigure, "State of the art of water chemistry of Japanese BWRs", Proc. Chem. In Water React. 94, p7, 1995
3. Y. Uruma, Y. Hemmi, T. Baba, "Crud deposition behavior on Zry-2 fuel cladding under BWR condition", Proceedings of 1998 JAIF International Conference on Water Chemistry in Nuclear Power Plants, October, 1998
4. Y. Goto, T. Takano, T. Nihei, Y. Hemmi, H. Hirasawa, T. Yotsuyanagi, "Ultra Low Crud Control in Onagawa Unit 2", Proceedings of 1998 JAIF International Conference on Water Chemistry in Nuclear Power Plants, October, 1998