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RADIATION FIELD CONTROL AT THE LATEST BWR PLANTS - DESIGN PRINCIPLE, OPERATIONAL EXPERIENCE AND FUTURE SUBJECTS

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ABSTRACT

Improvements of operational procedures to control water chemistry, e.g., nickel/iron ratio control, as well as application of hardware improvements for reducing radioactive corrosion products resulted in an extremely low occupational exposure of less than 0.5 man.Sv/yr without any serious impact on the radwaste system, for BWR plants involved in the Japanese Improvement and Standardization Program. Recently, ^{60}Co radioactivity in the reactor water has been increasing due to less crud fixation on the too smooth surfaces of new type high performance fuels and to the pH drop caused by chromium oxide anions released from stainless steel structures and pipings. This increase must be limited by changes in water chemistry, e.g., applications of modified nickel/iron ratio control and weak alkali control. Controlled water chemistry to optimize three points, the plant radiation level and integrities of fuel and structural materials, is the primary future subject for BWR water chemistry.

INTRODUCTION

Occupational exposure at nuclear power plants is determined by the three factors:

- 1) the radiation level where major inspection and maintenance operations are carried out;
- 2) the work time for each maintenance operation; and
- 3) the number of personnel needed for each maintenance operation.

In order to reduce the radiation level, control of radioactive corrosion products is essential and the procedures for controlling corrosion products without any serious impact on the radwaste system have been applied for plant operation as well as design of plant systems and major hardware at BWRs involved in the Japanese Improvement and Standardization Program (JISP)¹. At the JISP BWRs, systematic applications of the procedures led to extremely low radiation levels which reduced occupational exposure to less than 0.5 man.Sv/yr.

As a result of analyzing current water chemistry data, it is found that ^{60}Co radioactivity in the reactor water at the JISP BWRs is again increasing with the application of high performance fuel and the pH drop caused by Cr anion being released from stainless steel structures and pipings. Changes in fuel and structural materials have some impact on corrosion product behavior, while improvement of the water chemistry for controlling radioactive corrosion products has some impact on behaviors of fuel and structural materials.

HISTORIC ASPECTS OF BWR WATER CHEMISTRY

Since the first Japanese BWR, Tsuruga-1, started commercial operation in 1970, a lot of experiences with water chemistry and radiation control in BWR plants have been accumulated. Some historic aspects of water chemistry and radiation control of BWRs are summarized in Table 1. The experiences can be divided into six periods and topics of greatest interest have changed from simply fuel integrity, structural integrity and occupational exposure to their combination.

Table 1 Historic aspects of water chemistry and radiation control in BWRs

Period	Major events	Water chemistry & radiation control
1 (Before 1975)	observation of fuel defects radioactive effluent increase	fission product removal
2 (1975 - 1980)	IGSCC occurrence occupational exposure increase	impurity control oxygen injection
3 (1980 - 1985)	Japanese Improvement and Standardization Program	dual condensate demineralizers low cobalt containing materials
4 (1985 - 1990)	BWR plants with low shutdown radiation level	Ni/Fe ratio control
5 (1990 - 1995)	⁶⁰ Co increase at every latest operating cycle	advanced Ni/Fe ratio control
6 (After 1995)	to establish a trio of water chemistry requirements	controlled water chemistry (Ni/Fe ratio & pH control, HWC)

During the first period, poor water chemistry often caused defects on fuel claddings. Some impurities, e.g., suspended iron crud, deposit on the fuel surfaces to form thick deposit layers, which not only depress reactivity of the core but also prevent sufficient heat transfer from the fuel to the coolant which increases temperatures at the fuel surface and subsequently enhances the corrosion rate. In order to improve fuel integrity, suppression of crud concentration in the reactor water was applied along with structural improvements, e.g., moderation of mechanical interactions between cladding and UO₂ pellets, and operational improvements, e.g., pre-conditioning for densification of UO₂ pellets.

The major target in the second period was IGSCC of stainless steel pipings. Some impurities in the reactor water, e.g., chloride ion, enhanced IGSCC of stainless steel pipings. Concentrations of radiolytic species, e.g., H₂O₂, OH, and O₂⁻, should be suppressed to moderate corrosive circumstances, while those of metallic and organic impurities, which also enhance corrosion, should be controlled. In order to avoid IGSCC of the piping, suppression of chloride concentration and conductivity in the reactor water were applied along with developments of IGSCC resistant materials, e.g., low carbon containing stainless steel, and residual stress improvement. During this period, inspection and maintenance operation of primary pipings caused high levels of occupational exposure, which hindered effects to win public acceptance of nuclear power plants. Then,

lowered exposure become the main target for the third period.

Small amounts of corrosion products released into the cooling water, such as ^{60}Co , are activated in the core, becoming radioactive. Some of these products deposit on the walls of the recirculation pipings and their components, which causes shutdown doses around the primary cooling system, and then, radiation exposures of personnel working on inspection and maintenance around the primary system. In the third period, the desire for reduced occupational exposures required much severer criteria for water chemistry, particularly radioactive corrosion product control. The primary procedures for reducing corrosion products, the effects of which were evaluated on the shutdown dose rate, were incorporated into the Japanese Improvement and Standard Program (JISP), the details of which are described in the following section.

In the fourth period, Fukushima Daini-2, which was the first BWR designed and constructed through involvement in the JISP, started its commercial operation. In spite of its application of radiation reduction procedures too low iron crud concentration caused an increase of ^{60}Co radioactivity in the reactor water, which was moderated by assistance of additional water chemistry control. ^{60}Co in the reactor water could be successfully reduced by adding suitable amounts of iron crud in the feed water and enhancing redeposition and fixation of ^{60}Co as cobalt ferrite at fuel surface. In order to suppress insoluble ^{60}Co radioactivity in the reactor water, the added iron crud should be controlled to keep a suitable nickel/iron ratio (Ni/Fe ratio < 0.5) of minimum amounts to form CoOFe_2O_3 and NiOFe_2O_3 at the fuel surface.

Recently, ^{60}Co radioactivity in the reactor water at the JISP BWRs is increasing due to less crud fixation on the too smooth surfaces of the new type high performance fuels and to the pH drop caused by chromium oxide anions released from stainless steel pipings. Presently, in the fifth period, attempts are being made to decrease ^{60}Co radioactivity by applying weak alkali control and improved Ni/Fe ratio control.

Water chemistry is affected by behaviors of fuel and structural materials, while fuel and structural materials are also affected by water chemistry. In the future's sixth period, establishment of "controlled water chemistry" will be the goal which will satisfy a trio of requirements, shutdown radiation reduction, integrity of fuel cladding and integrity of structural materials.

JAPANESE IMPROVEMENT AND STANDARDIZATION PROGRAM

The basic design concept of the JISP BWRs is characterized by the following features.

- a) High reliability: duty factor of 70%.
- b) Minimum shutdown period for scheduled refueling and annual maintenance: 85 days.
- c) Minimum occupational exposure: 1.3 - 1.5 man.Sv/yr.

Occupational exposure is determined by three factors 1) - 3) listed in the Introduction. The basic concepts and main procedures for reducing occupational exposure through these three factors are shown in Table 2.

In order to reduce the radiation level, control of radioactive corrosion products is essential. Procedures with sufficient effects and high reliabilities have been systematically applied in JISP BWRs, and efforts have also been made to reduce the radwaste sources, which accompany the increase of corrosion product removal. A schematic diagram for radioactive corrosion product behavior is presented in Figure 1. The process for

radioactive corrosion product behavior are divided into three steps, e.g., generation, activation and deposition, where suitable procedures for controlling corrosion products are applied. The procedures for reducing shutdown dose rate applied in JISP BWRs are shown in Figure 2.

Occupational exposure at the JISP BWRs during the first refueling and annual inspection periods can be reduced to less than 0.5 man.Sv/yr by applying suitable Ni/Fe ratio control (Figure 3).

Table 2 Basic concepts and main procedures for reducing occupational exposure

Basic concepts	Shutdown radiation reduction	Improvement of personnel mobility	Automation for inspection & operation
Main procedures	(1) Low cobalt containing materials (2) Oxygen injection into feed water (3) Prefiltration of condensate water (4) Feed water recirculation system	(1) Application of improved Mark II type PCV*1 i) Increasing operation space ii) Improvement of carriage path	(1) Automatic ISI*2 machine (2) Automatic CRD*3 exchanger (3) Automatic fuel assembly exchanger (4) Improved LPRM*4 exchanger

*1 pressure containment vessel

*2 in-service inspection

*3 control rod drive

*4 local power range monitor

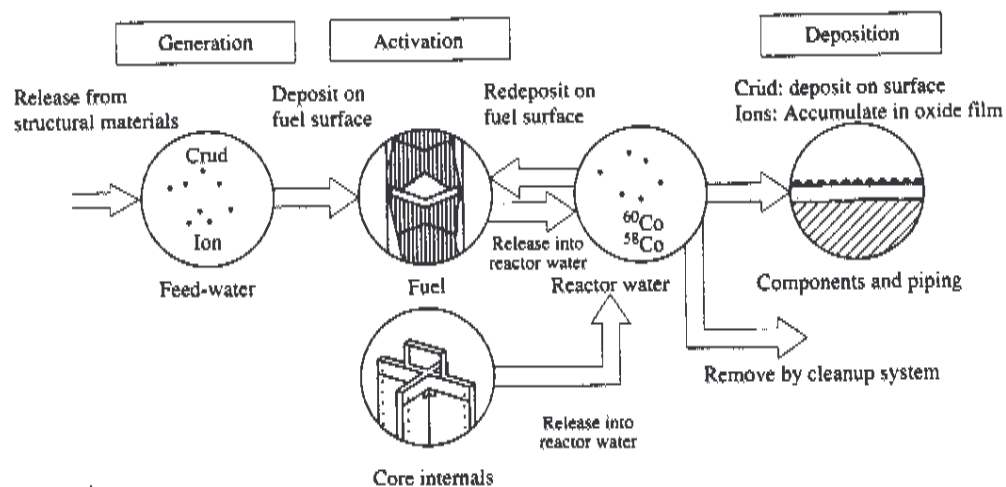


Figure 1 Radioactive corrosion product behavior

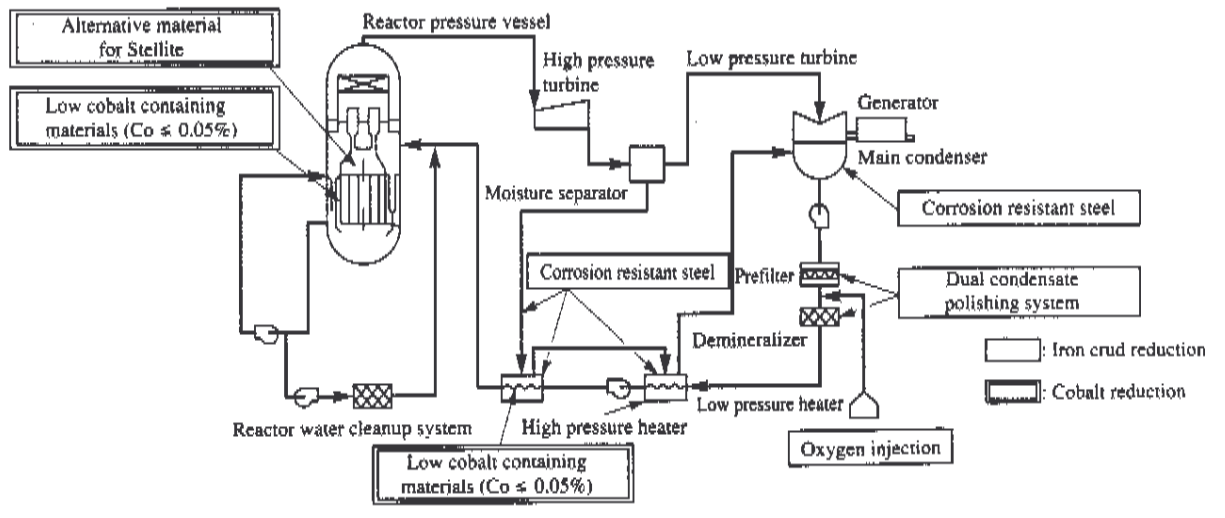


Figure 2 Application of procedures for reducing shutdown dose rate

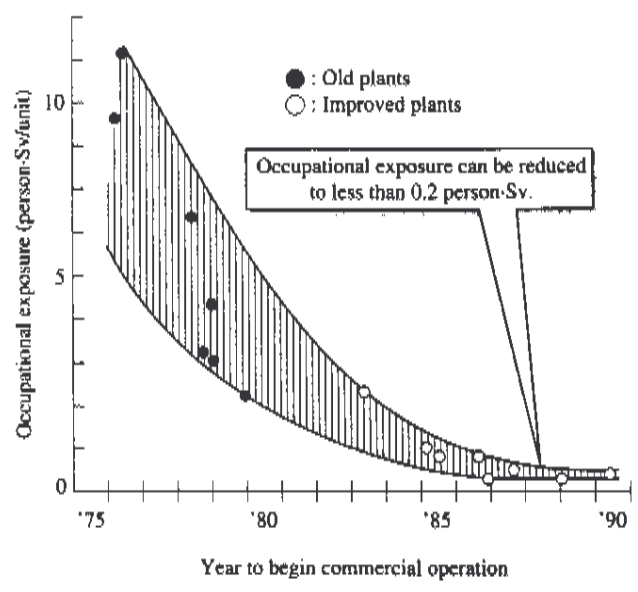


Figure 3 Occupational exposure during first refueling and annual inspection periods

EXPERIENCES WITH WATER CHEMISTRY OF THE LATEST BWRs

The relationship between shutdown dose rate and occupational exposure is shown in Figure 4. With a few notable exceptions, occupational exposure is in proportion to the shutdown dose rate, which is also proportional to ^{60}Co radioactivity in the reactor water. The average ^{60}Co radioactivity in the reactor at each operational cycle of several JISP BWRs is shown in Figure 5. Some values for the latest cycle exceed the target value of 2 Bq/ml necessary to keep a shutdown dose rate of less than 0.5 mSv/h. Increased ^{60}Co radioactivity seems to be caused by changes in water chemistry and fuel.

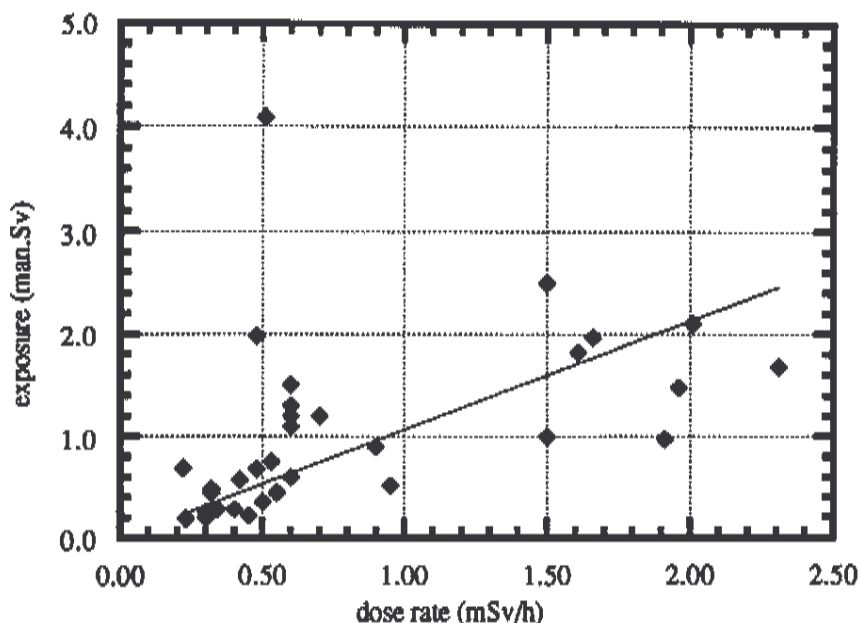


Figure 4 Relationship between shutdown dose rate and occupational exposure

One of the typical changes in water chemistry in the latest BWRs is decreasing pH in the reactor water². Increasing chromium anion concentration decreases pH in the reactor water, making it slightly acidic (6.6 - 6.8). This enhances ^{60}Co release from deposits on the fuel surface which in turn increases ^{60}Co radioactivity in the reactor water. The relationship between ^{60}Co radioactivity and pH in the reactor water is shown in Figure 6. Although ^{60}Co radioactivity is affected not only by pH, but also by concentrations of iron curd and cobalt, Figure 6 shows that rather high pH causes low ^{60}Co radioactivity, while low pH causes rather high radioactivity. As a result of injecting sodium hydroxide in the reactor water for weak alkali control, the contribution of Cr anion to pH is moderated and then pH increases to moderate the ^{60}Co radioactivity buildup.

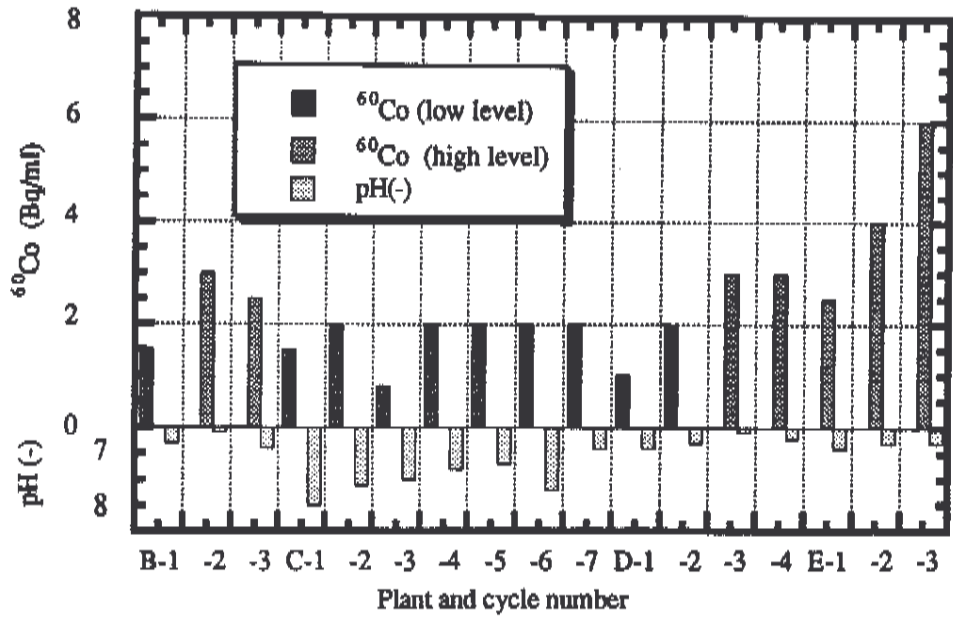


Figure 5 Relationship between ^{60}Co radioactivity and pH in the reactor water at each operational cycle of four BWR plants

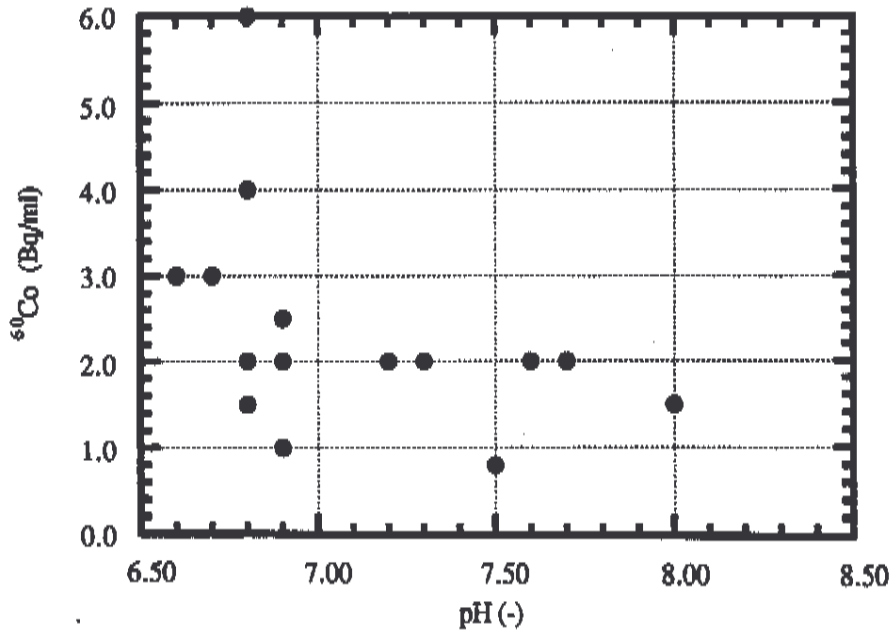


Figure 6 Relationship between ^{60}Co radioactivity and pH in the reactor water

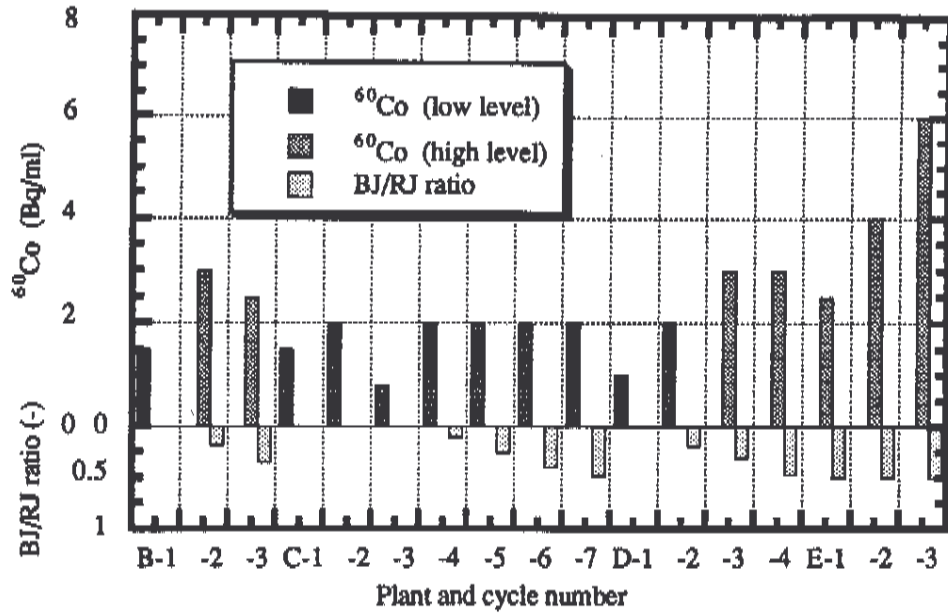


Figure 7 Relationship between ^{60}Co radioactivity and BJ fuel load factor in the core at each operational cycle of four BWR plants

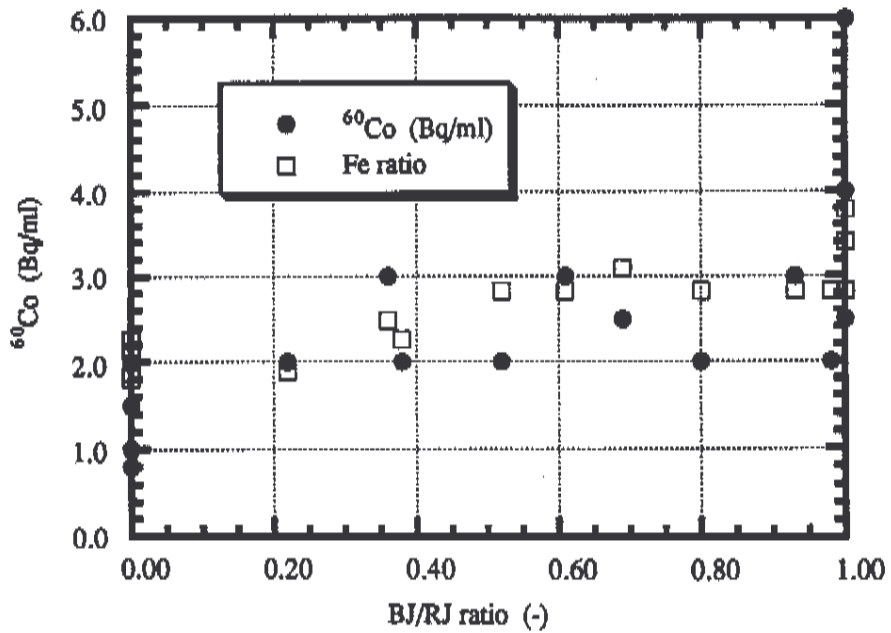


Figure 8 Relationship of ^{60}Co radioactivity and Fe ratio of [real crud] to [ideal crud] to the BJ fuel load factor

Another change in water chemistry is iron crud concentration in the reactor water². As a result of applying new type high performance fuel (designated as BJ fuel) with mechanically polished smooth surfaces, the deposition rate of crud on them is reduced, which causes some delay in fixation of cobalt as cobalt ferrite to increase ⁶⁰Co radioactivity in the reactor water (Figure 7), even if enough iron crud is supplied to satisfy a suitable Ni/Fe ratio. Iron crud comes mainly from the feed water line, while it is removed mainly at the fuel surface. It is easy to estimate crud concentration in the reactor water by using crud concentration in the feed water and the crud deposition coefficient at the fuel surface. As a result of applying the high performance fuel, the iron crud concentration in the reactor water is higher than the estimated values.

The ratio of measured iron crud concentration (the real concentration) to the calculated ones (the ideal concentration) is proportional to the load factor of BJ fuel in the core as shown in Figure 8. Once deposited on the fuel surface, iron crud is easily released into the reactor water, and then repeats of deposition and release follow to cause a high concentration in the reactor water. Cobalt deposited on the fuel surface missing its partner to form cobalt ferrite, and there is some delay for fixation to occur which increase ⁶⁰Co radioactivity in the reactor water. The relationships between ⁶⁰Co radioactivity or the ratio of real crud concentration to the ideal one, to the BJ fuel load factor are shown in Figure 8. The data support the contributions of BJ fuel application to increasing ⁶⁰Co radioactivity.

SHORT TERM COUNTERMEASURES FOR RADIATION REDUCTION

In a few years, water chemistry control will be focused on reduction of radioactive corrosion product accumulation, so as to avoid any effects of changes in fuel cladding and structural materials.

In order to reduce ⁶⁰Co radioactivity, weak alkali control (pH 7.0 - 8.0) is being applied to moderate ⁶⁰Co release from fuel surfaces, while improved Ni/Fe ratio control, in which much iron crud is supplied at an early stage to cover the fuel surface completely with crud and thus improve the crud deposition rate, is also being applied to enhance fixation of ⁶⁰Co on the fuel surface as cobalt ferrite².

LONG TERM COUNTERMEASURES FOR CONTROLLED WATER CHEMISTRY

Cooling water, fuel claddings and structural materials should be optimally selected to satisfy a trio of requirements, reduction of shutdown radiation level, integrity of fuel cladding and integrity of structural materials. Water chemistry should be controlled to improve performances of fuel cladding and structural materials, while fuel cladding and structural materials should also be moderated to improve water chemistry. A combination of Ni/Fe ratio control, weak alkali control and fuel cladding modification for radiation reduction has been proposed.

"Controlled water chemistry" seems to be the major route in the future which will allow the three requirements to be satisfied through reliable and easy chemical procedures and with less influences of changes in plant operation procedures and structural materials³⁻⁴. Severe water chemistry control is necessary, but some of the target values for the different requirements conflict with each other and optimal target values must be determined

to balance them. In order to establish "Controlled Water Chemistry," first effects of major water chemistry factors on materials and corrosion product behaviors at elevated temperatures under radiation (and under the reactor water condition) should be quantitatively evaluated, from which the mechanism determining the effects can be derived. It is important to control water chemistry by evaluating the current plant status as well as predicting long term effects on materials.

CONCLUSIONS

Water chemistry control with mono-purpose optimization for reduction of occupational exposure has been established by evaluating actual plant operational data in JISP BWRs with lower shutdown radiation levels. In the future, "water chemistry control" with tri-purpose optimization can be established by confirming fundamental data of water chemistry and materials and then estimating plant future trends.

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Author Biography

Shunsuke Uchida is a Senior Chief Researcher and Head of the Reactor Preventive Maintenance Research Center at Energy Research Laboratory, Hitachi, Ltd. He takes the leadership of research and development (R&D) programs concerning BWR water chemistry and structural materials. Before joining the R&D programs, he worked for radiation shielding and protection at BWRs and LMFBs, and was a visiting exchanger with the Neutron Physics Division of Oak Ridge National Laboratory. He has a B.Sc. in Physics from Osaka University (1964) and a Ph.D. in Nuclear Engineering from the University of Tokyo (1979). He won three Zirconium Medal Awards of the Atomic Energy Society of Japan and a Ministerial Award of Science and Technology Agency for outstanding contributions to the research of BWR water chemistry.

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DISCUSSION**

Unknown: I have a collective question. I know that your major enemy is cobalt-60, but I would like to know the different strategies in the different countries dealing with fuel failures in BWRs. Do you perform a major shutdown, or do you wait for a plant outage? If you didn't, have you an idea of the evolution of the dose rates of the alpha contaminations with the activity of neptunium or fission products?

Uchida: Fortunately, for several years we have not had any serious fuel damage. Fuel defects are caused by some impurity of coolant, but we have very few experiences of fuel leakage, so we don't worry about this contamination of neptunium or the fission products. We only think only about cobalt-60 for determining the shut-down dose rate.