REDUCTION OF RADIATION EXPOSURE IN JAPANESE BWR NUCLEAR POWER PLANTS

Yoshitake Morikawa
Toshiba Corporation
Applied Metallurgy & Chemistry Department
ISOGO Nuclear Engineering Center
8 Shinsugita-Cho, Isogo-ku
Yokohama, Japan

INTRODUCTION

The reduction of occupational exposure to radiation during the annual inspection and maintenance outages of Japanese boiling water reactors (BWR) is one of the most important objectives for stable and reliable operation.

It was shown that this radiation exposure is caused by radionuclides, such as Co-60, Co-58 and Mn-54 which are produced from the metal elements Co, Ni, and Fe present in the corrosion products of structural materials that had been irradiated by neutrons. Therefore, to reduce radiation sources and exposures in Japanese BWRs, attempts have been reinforced to remove corrosion products and activated corrosion products from the primary coolant system. This paper describes the progress of the application of these measures to Japanese BWRs.

Most Japanese BWR-4 and BWR-5 type nuclear power plants started their commercial operations during the 1970s. With the elapse of time during operations, a problem came to the forefront, namely that occupational radiation exposure during plant outages gradually increased, which obstructed the smooth running of inspections and maintenance work. To overcome this problem, extensive studies to derive effective countermeasures for radiation exposure reduction were undertaken, based on the evaluation of the plants' operation data.

In particular, the Improvement and Standardization Program to establish Japanese Light Water Reactors, which aimed at improving plant reliability and availability and at reducing occupational radiation exposure, was established after 1975 under cooperative efforts by the Japanese electric power companies and plant manufacturers.

Following this program, a series of countermeasures also were applied to the older BWRs which had started their commercial operations before the Improvement and Standardization Program was adopted.

Historically speaking, all the Japanese BWRs of the initial generation, the so-called older BWRs, gave unexpectedly high occupational radiation exposures that increased annually during their refueling and maintenance outages. After several years of effort to clarify this problem, it became clear that the occupational exposure was determined by radioactive corrosion products, particularly Co-60 and Mn-54 deposited on major components and pipings of the primary coolant system, and that the following factors have a big influence on the buildup of radionuclide and, consequently, on occupational radiation exposure:[1]

1. input of iron crud into the reactor water from the feedwater (concentrations of Fe crud in the feedwater),
2. cobalt contents in structural materials, especially in-core materials,
3. capacity of the reactor's water cleanup system,
4. quality of the reactor water, and
5. condition of the inner surface of pipings and components.

Based on these findings, a wide variety of countermeasures were developed and proposed to reduce and control the radiation dose level around the primary coolant circuits (Figure 1). A detailed breakdown of these measures is shown in Table 1; most of them already had been adopted by both older and newer plants, achieving excellent BWRs with very low occupational radiation exposures.

Now, during this decade the water quality and radionuclide concentrations in the reactor water of all the Japanese BWRs changed greatly, and there are new problems which have to be solved to further reduce radiation exposure.

In this paper, I discuss some of the typical measures which were proven to be effective for reducing radiation dose rate in older and new Japanese BWRs from the standpoint of the improvements in materials, systems, and operations, and the results from adopting these measures. Some of the new items being studied in Japan also will be presented.

MATERIAL IMPROVEMENT

Sampling Line Material

In advancing the studies on the behavior of corrosion products, it is very important and fundamental to obtain accurate data on water chemistry for the operating plants. It was found that the conventional stainless-steel sampling line for sampling water at high temperatures gave incorrect values for the concentration of corrosion products, especially for Co and Ni due to the contamination of corrosion products released from materials of the sampling line.

Titanium (Ti) was found to be the most appropriate material for the high-temperature sampling; consequently, conventional sampling line tubings and valves for the final feedwater sample made of stainless steel and Stellite were replaced by titanium ones in most Japanese BWRs.

Reduction of Iron Crud

Most of the iron (Fe) crud fed into the reactor comes from feedwater. Its origin in the final feedwater is the crud which was generated upstream of the main condenser and leaked through the condensate treatment system. Iron crud also comes from corrosion products generated from the components of the feedwater system.

Low alloy steels, STPA-23 and A387Gr.11 (1.25 Cr-0.5 Mo steel), were used to reduce corrosion-erosion in extraction steam pipings and their drain pipings. These materials were selected from laboratory loop tests.[2] Newer BWR plants also chose these alloys for the moisture separator and its drain pipings. Even in older plants, the same materials were adopted, in part, for the equipment and pipings of the condensate and heater drain systems.

For the material of main condenser, a special carbon steel which contains small amounts of chromium and copper, SMA-41 (0.3 Cr-0.3 Cu steel), was used in place of plain carbon steel after considering the results of inplant corrosion tests. Figure 2 clearly shows the effect of material replacement on reducing the concentration of Fe crud in condensate water is clearly seen.

Reduction of Cobalt

Cobalt is released into the primary cooling water from various parts of the primary system. Generally, cobalt is contained in stainless steel and in nickel-based alloys as an impurity of the nickel component, and also in cobalt base alloys. Therefore, cobalt can be reduced by using low cobalt materials in place of conventional structural materials. The materials to be replaced with low cobalt materials should be selected after an evaluation of the contribution of each material to Co-60 concentration in reactor water.

From this point of view, the first materials to be replaced by low cobalt materials should be 1) materials with a large surface area in contact with the high temperature primary water, such as stainless steel tubings of the feedwater heater system, 2) materials with a high cobalt content, such as Stellite, and 3) in-core materials which receive high neutron irradiation, such as springs made of Inconel in fuel bundles.

For example, the material used for pins and rollers on the control rods of conventional plants were made of the cobalt-base alloy, Stellite. Although the surface area of these parts is small, it meets the two of these conditions of high cobalt content and in-core materials, and so should be replaced with low cobalt alloys. All recent Japanese control rod pins and rollers are made of a newly developed cobalt-free alloy[3] instead of the cobalt base alloy, Stellite.

Inconel springs in the spacers of fuel bundles were evaluated as the biggest sources of Co-60 and Co-58 in reactor water because ordinary Inconel contains approximately 0.2% of element cobalt and they always receive high levels of irradiation from thermal and fast neutrons. Consequently, all Japanese plants recently changed to Inconel springs of low cobalt content (<0.05%).

Low cobalt stainless steel in which the cobalt content is less than 0.05% was recently used for feedwater heater tubings in newer plants instead of unspecified ordinary stainless steel. In the newer plants, some of the reactor's internal equipment also was replaced by this low cobalt stainless steel.

Figure 3 shows how the extent of the influence of each component as a source of Co-60 has changed, from conventional plants with low cobalt materials to newer plants that have incorporated them to a great extent.[4] Assessments showed that Co-60 sources were reduced to about one fourth by the adoption of low cobalt and cobalt free materials.

From this evaluation, it was found that the main sources of Co-60 consist of those generated from Stellite in valves. Therefore, it is most important that new, cobalt-free alloys are developed for the hard-facing materials of valves, especially for large bore valves.

Titanium Condenser

As shown in Table 1, titanium was adopted for the tubing material in main condenser of newer plants to improve the efficiency of the turbine heat exchanger and the quality of condensate water.

The feedwater of the plants which adopted a Ti condenser, in addition to using a system called a dual condensate treatment system, showed quite excellent water quality, approximately 0.06 μS/cm. Consequently, non-chemical regeneration operation of the condensate demineralizer was unnecessary for more than five years.

One of the desirable influences of this improvement was that the generation of secondary radwaste was reduced to a large extent. Another one, the most important one from the viewpoint of control of radiation buildup, was that the impurities in reactor water were steadily maintained at very low level, around 0.1 μS/cm. This suggests that the buildup rate of radioactivities on out-of-core pipings and equipment is controlled at desirable levels.

Pretreatment of Material Surface

Pretreating the material surface to reduce buildup of activity by controlling the characteristics of its oxide film is considered as one effective measure among other remedies. Factors which contribute to the rate of deposition of activity by pretreating materials are the smoothness of the material's surface, protectiveness of the oxide film, thickness of the oxide film, and so forth.

One pretreatment method, prefilming by dissolved oxygen, was employed in several plants. The first experience of prefilming for the recirculation pipings was carried out at Kashiwazaki-Kariwa No. 1 unit (K1). In K1, the prefilming was carried out before the commissioning test. The concentration of oxygen in the reactor water was kept about 300 ppb by dosing oxygen gas into the water of the control rod drive system. The heat source was Joule heat from running the recirculation pump. Radiation suppression rate was about 15% determined by the coupon test during the first cycle operation of K1.

Many different kinds of pretreatment methods have been developed worldwide. Figure 4 shows an example of the results of those studies. Two sorts of coupons, one group of which consisted of as-received coupons relative to ordinary recirculation piping, and other coupons which were mechanically and electrolytically polished, were exposed in the reactor water of four operating plants. The figure shows that electropolishing the recirculation pipes could control the radioactivity buildup to less than one half in comparison with unpolished ones.

Our recent efforts focused on developing techniques to reduce the dissolution of ionic radioactivities from in-core materials, particularly Inconel springs in the fuel spacers. After many kinds of pretreatment studies, including oxidation by chemical treatment, high-temperature oxidation in air was chosen as the most realistic technique from laboratory tests. Figure 5 shows the trends in metal release from a coupon oxidized by conventional treatment and a coupon in high-temperature air when tested in BWR water. The rate of release of the metal was very small for the latter. After this test, the distributions of metal oxide in the oxide layers were determined, as shown in Figure 6. It seems that the excellent protective properties of the air-oxidized coupon against corrosion came from the high contents of nickel ferrite and chromium oxide in the oxide film.[5]

SYSTEM IMPROVEMENT

Various kinds of improvements to systems to reduce radiation exposure during each maintenance and inspection outage were proposed from the viewpoints of both reducing radiation dose-rate and saving working time around the radiation fields. Table 1 shows some improvements that have been applied to the older plants, newer plants, will be used in future plants.

Improvements in the Water Treatment System

Generally, the efficiency of crud removal by a mixed-bed type, cation/anion ion-exchange beads resin in a condensate polishing system were thought to be insufficient to reduce Fe crud concentration in feedwater below 1 ppb. Through observations of water chemistry during condensate demineralizer operations, it was found that cation resin, especially aged cation resin, removed not only crystallized iron oxide but even less well crystallized iron compounds generated from the material surfaces upstream of the condensate system, as shown in Figure 7.[6,7]

Based on studies of the properties of aged cation resins, several new cation resins were developed and tested in actual condensate water, one of which showed an excellent ability for crud removal (Figure 8). At present, the mechanical properties of other improved cation resins are being tested, using condensate water from an operating plant.

Most Japanese 1100MWe-class BWRs employ a dual condensate treatment system to reduce Fe crud input into the reactor. The concept of dual condensate treatment system is a purification system consisting of a prefilter to remove Fe crud in condensate water and a demineralizer to eliminate impurities during occasional sea-water intrusion. The prefilters used before the condensate demineralizer are classified into two types, powdered resin-type water polishers, and hollow-fiber filter systems (HFF).

However, some technical problems with powdered resin-type filters were revealed, including the efficiency of crud removal and shortening of run length. Typical decontamination factors and run lengths of powdered resin type filter ranged between 3 to 7 ppb, and 10 to 25 days, respectively. To solve these problems, several countermeasures were proposed and tested. Recently, some countermeasures, shown in Table 2, were proved effective by in-plant tests at operating plants; in the best case a very long run length of about 200 days was achieved (Figure 9).

The other improvement was the adoption of newly developed Hollow Fiber Filter (HFF) system upstream of the deep bed demineralizer. This system was first applied to a radwaste treatment system, and then to the conventional plants to save outage time by shortening the re-startup cleaning time of condensate and feedwater. For the latter purpose, a HFF having the capacity of 30% feedwater flow was first applied to Fukushima Daiichi Nuclear Power Station No. 3 Unit (1F-3); after operating successfully during restartup of the plant, it was put in-service to purify the condensate water during steady state power operation.

The efficiency of crud removal was measured by the iron species contained in the condensate water. A result, shown in Figure 10, gives measurements for a precoat-type filter. HFF had an excellent performance, even for amorphous iron species; Figure 11 shows the efficiency of Fe crud removal measured for condensate water of 1F-3 plant. Hollow fibers used in the 1F-3 plant initially were hydrophobic; later ones were improved and had hydrophobic properties, tending to have a longer module life and being easier to handle. It was proved by in-plant tests that the hydrophilic hollow fiber had smaller increase in differential pressure compared to the hydrophobic one. This hydrophilic filter module has been used in 1F-3 in place of a hydrophobic module after four successful years in operation.

Now, the HFF system has been adopted by many Japanese BWRs, and replacement of powdered-resin type condensate polishers by HFF system is also discussed because of its better crud removal performance and its substantial reduction of secondary radwaste generation.

Shielding Radiation Sources in the Drywell

To attain a low dose-rate for the working areas in the drywell during maintenance and inspection outages, shieldings were applied to the main radiation sources from the recirculation and reactor water cleanup (RWCU) pipings of conventional plants and new plants. Figure 12 shows an example of the savings in radiation exposure by using shieldings for the radiation-contaminated pipings in drywell. This has become one of routine remedies for reducing radiation dose-rate in Japanese BWRs.

Minimization of Radiation Dose Rate in the RWCU System

The reactor water cleanup (RWCU) system has been manufactured with thin- walled pipes, adequate to withstand the inner pressure, but not thick enough to function as a shield against radiation deposited on its inner surface area.

Some improvements for reducing radiation dose rate reduction in this system were carried out in the newer plants. The RWCU pumps were relocated from a high-temperature area to a low-temperature area downstream of the nonregenerative heat exchangers; the temperature dependency of activity deposition on carbon steel (used in Japanese BWRs).

The second improvement of this system was shortening the pipe length by rearranging the piping routes in the drywell. The third improvement was shielding part of the RWCU pipings in the drywell.

OPERATIONAL IMPROVEMENT

Ni/Fe Ratio Control Operation

In newer Japanese BWR plants, it has become easy to decrease the input of Fe crud into the reactor water by using the dual condensate polisher. Therefore, at first it was thought that it would be easy to keep crud radioactivities such as Mn-54, Fe-59, Co-58, and Co-60 at low levels.

However, in one plant, an adverse phenomenon on the radioactivities in reactor water was observed; iconic Co-60 and Co-58 in the reactor water in the presence of a small amount of Fe crud was strongly effected by the ratio of Ni/Fe input. That is, to suppress ionic Co-60 and Co-58 concentrations in reactor water, a little more than two times the amount of Fe crud compared to nickel should be fed into the reactor water. The results of several experiences in controlling the Ni/Fe concentration ratio are shown in Figure 13.

A key process may be the reaction between Ni and Fe on the fuels' surface:

$$Ni^{2} + Fe_2O_3 + H_2O = NiFe_2O_4 + 2H^+$$

Iconic cobalt also reacts with Fe, forming cobalt ferrite. Precipitation of many kinds of iron oxides with ionic Ni, Co-58, and Co-60 were carried out in laboratory experiments in the presence of trace amounts of Ni and Co to simulate the reactor water environment. Iron oxides which were not highly crystallized reacted faster with ionic species to form Ni ferrite and Co ferrite compared with crystallized iron oxides, such as hematite and geothite.[8]

Taking these findings into consideration, measures to control and lower iron concentrations in the feedwater were applied to crud chemistry in newer BWR plants. These measures included iron crud dosing with a partially bypassed flow-condensate prefilter, and an Fe crud dosing system installed at the feedwater system. The optimum Fe concentration in feedwater appear to range from 0.2 to 0.5 ppb. Crud control in newer BWR plants is operated carefully, with this target of Fe crud concentration in the feedwater, so keeping radiation levels low.

Suppression of Radionuclides during Shutdown Operation

It is well known that radioactive crud concentration in the reactor water becomes higher during shutdown operations than during normal power operations, sometimes increasing by more than tenfold. This phenomenon is thought to be caused by the release of part of the fuel crud into the reactor water; this crud has a higher specific radioactivity than that of crud in the reactor water during normal power operation.

Some shutdown procedures to minimize the above increase were studied in several Japanese BWRs, and the following procedures were effective and realistic; the cooling rate of the reactor water was reduced to less than 15°C/h, and the reactor pressure was held constant for 3 to 4 hours at $50kg/cm^2$. The results showed that the modified shutdown procedures suppressed the maximum radioactivity concentration in the reactor water by one to two orders of magnitude compared to that from conventional practices.

Integrated Layup Practices

The adoption of integrated layup practices is thought to significantly reduce the amount of corrosion products in feedwater. Without layup practices, high crud loadings were observed on the condensate demineralizer when the plant was started up after refueling outages, particularly in older BWRs.

It was expected that cleaning the condenser and hotwell could remove considerable quantities of crud and prevent its input to the reactor vessel at conventional plants which used only a condensate demineralizer. From this point of view, as mentioned before, HFF was adopted by conventional plants.

If a HFF system having 30% capacity of the feedwater flow was installed upstream of the condensate demineralizer, the cleaning time necessary to obtain the target value of 200 ppb in the final feedwater was only two days compared in two weeks with only a condensate demineralizer at the same plant. In the case of the dual condensate polisher system, only prefilters were put into service for the purpose of pre-restartup flushing of feedwater and condensate water.

EFFECT OF RADIATION CONTROL MEASURES

All measures developed so far to reduce radiation dose rate, some important ones of which were discussed in the preceding sections, were adopted by the newer Japanese BWRs from their initial designing stage. Many of them also were applied to older, conventional plants. As the result of these measures, the general area dose rates in the drywell of newer plants are kept comparatively low, as a typical example shows in Figure 14.

This low dose rate, combined with efforts to reduce exposure time, have resulted in a record of low radiation exposure. Older, conventional plants also show noticeable decreasing trends in radiation exposure after adopting many of these measures.[9] Figures 15 and 16 give typical examples of radiation exposures of both older and newer BWR plants.

FURTHER REDUCTION IN RADIATION EXPOSURE

Older Plants

Although the radiation dose rate and, hence, the radiation exposure in older plants has decreased by adopting many of the countermeasures developed so far, it is not yet satisfactory. To find further remedies, extensive measurements to determine the amounts and morphologies of radionuclides deposited on the inner surfaces of various pipings and equipments were carried out in a typical old plant. The measurements revealed unexpectedly large amounts of radioactive soft crud or slightly adhered insoluble crud still existing, even on vertical pipings. Figure 17 shows the result of calculating the relation between radiation exposure and radiation sources in the drywell;[10] about 30% of the radiation exposure comes from the soft crud in the drywell, which means that a considerable amount of radiation exposure could be easily reduced by mechanical cleaning such as water-jet flushing.[11]

Newer Plants

Although the occupational radiation exposures of newer plants were very low at the first refueling and maintenance outages (Figure 18), they have shown a gradual increase year by year. It seems clear that this increase comes from Co-60 buildup on the material surface of the primary circuit (Figure 18). Two remedies are considered to mitigate this buildup; one is to further reduce Co and Co-60 sources by measures such as replacing the Stellite in valves, and using air-prefilmed Inconel for fuel spacer springs as described before, and the other is to develop methods to mitigate the buildup rate of Co-60. For this purpose, a more detailed and basic knowledge than we have now is needed about the interaction between oxide film and ionic metal elements.

CONCLUSION

The radiation control measures adopted in Japanese BWRs were qualified by the investigating data on operating water chemistry and radiation levels. Following the Improvement and Standardization Program established by electric power companies and plant manufacturers, the occupational radiation exposure was reduced this decade by factor of more than ten. Standing on the ALARA concept, however, more measures need to be developed to further reduce dose rate and radiation exposure.

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AUTHOR BIOGRAPHY

Yoshitake Morikawa is a Chief Specialist at the Applied Metallurgy and Chemistry Department, Nuclear Energy Division, Toshiba Corporation in Japan. He is working in the field of water chemistry related to radiation control technology to apply dose rate reduction to plants and candidate water chemistry control (Hydrogen Water Chemistry) of BWR Plants. Mr. Morikawa is also responsible for the promotion and management of the research and development of water chemistry studies at the Nuclear Energy Division in his company. He has a B.Sc. in Chemistry form Gamma University, Gamma Prefecture, Japan.

Toshiba Corporation
Applied Metallurgy & Chemistry Department
ISOGO Nuclear Engineering Center
8 Shinsugita-Cho, Isogo-ku
Yokohama, Japan

Phone: +81 045 770 2039
Fax: +81 045 770 2355