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Highlights of EPRI Radiation Exposure Management Program

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Abstract

Radiation exposures at U.S. nuclear power plants continue to decline, but radiation protection engineers face increasing challenges as a result of shorter outages, core uprating and remedial measures to mitigate materials degradation. This paper describes recent technology advances that have been implemented to control out-of-core radiation dose rates.

The use of noble metal chemical application to mitigate core internals cracking in BWRs resulted in increase in radiation fields at some plants as a result of redistribution of activated corrosion products. This paper describes the investigation of corrosion transport processes that led to successful recommendations to control fields.

Zinc injection has been implemented at several PWRs, resulting in ~20% reduction in radiation fields per cycle. This paper outlines work to optimize zinc injection to maximize the dual benefits of reduced stress corrosion cracking and radiation control, while avoiding adverse side effects.

A patented technique for the removal of activated corrosion products from fuel cladding using ultrasonic cleaning has been implemented at several PWRs, and is currently being qualified for BWR applications. The latest plant data concerning this technology are presented.

The EPRI Radiation Field Control Manual provides RP managers, engineers, and chemists with a valuable reference to the current field control and reduction technologies that are employed in the United States nuclear power plants. This paper discusses briefly the material described within the document.

I. INTRODUCTION

Occupational radiation exposures at light water reactors worldwide have decreased in a trend extending over two decades. Median collective exposures at US plants and electric generation data, shown in Figure 1, indicate that the collective exposure per MW electricity generated has dropped by an order of magnitude in the last 20 years. Despite this outstanding success story, challenges remain, as plants age, output is increased and outages become shorter. Accordingly, EPRI has actively pursued advanced technology to reduce out-of-core radiation fields, with recent developments described in this paper.

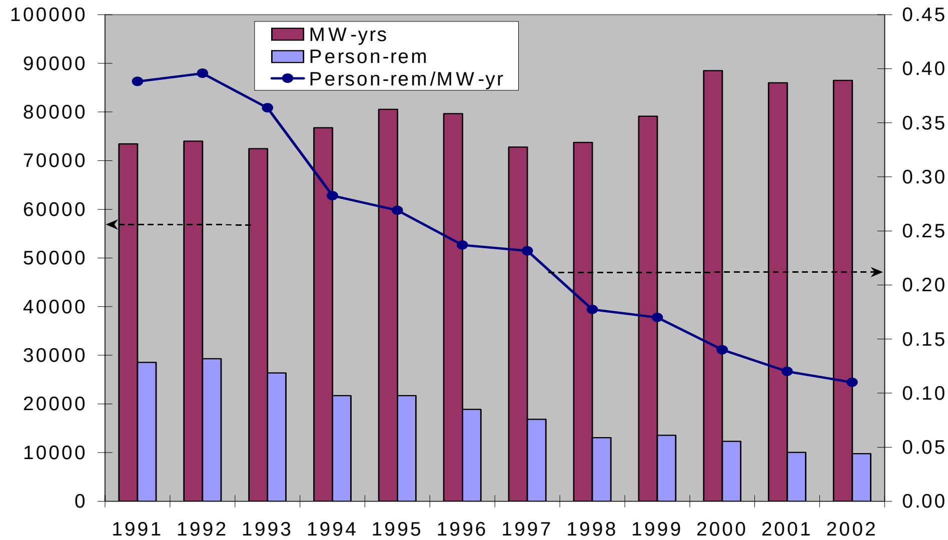

Figure 1: Collective radiation exposure and electricity generated at US nuclear power plants

II. BWR NOBLE METAL CHEMICAL APPLICATION

BWRs implement hydrogen water chemistry to reduce electrochemical potential and mitigate cracking of core internals. In the 1990's, hydrogen concentrations were increased to provide more protection in the reactor vessel. This chemistry change resulted in increased out-of-core radiation fields; most plants introduced depleted zinc injection to control dose rates that increased from the increased hydrogen rates.

Main steam line radiation from N-16 activity increases under HWC conditions because the nitrogen species formed under reducing conditions are more volatile than in oxidizing environments. The magnitude of the effect increases at higher hydrogen concentrations. Studies at General Electric showed that the presence of noble metals on structural materials would significantly reduce the hydrogen concentration required to achieve the IGSCC protection potential of -230mV(SHE). Noble metal chemical addition (NMCA) was introduced at Duane Arnold BWR as an in-situ method of reducing the amount of hydrogen required to lower the ECP on material surfaces, which would also mitigate the effects on operating radiation fields.

One method of achieving the ECP specification, Noble Metal Chemical Application (NMCA), was developed to avoid increased dose rates and high hydrogen usage. NMCA deposits very small amounts of platinum and rhodium metal on the wetted surfaces within the reactor vessel and reactor coolant system. These noble metal deposits catalyze recombination reactions of hydrogen with O_2 and H_2O_2 at these surfaces. Protective ECPs are achieved when the molar ratio of hydrogen to total oxidant in reactor water reaches a value equal to or greater than two. In the BWR, the molar ratio reaches the value of two at very low feedwater hydrogen addition concentrations (usually between. 0.1 and 0.15 ppm). Also, there is little or no increase in main steam line radiation from N-16 activity at these hydrogen addition levels.

During the initial implementation of Moderate HWC to the BWR fleet, it was noted that the introduction of feedwater hydrogen significantly increased shutdown dose rates at some plants at the end of that fuel cycle. However, the shutdown dose rate effects of hydrogen water chemistry can be mitigated by feedwater zinc addition. Similar effects are found with NMCA.

In the first cycle after NMCA application, all plants have seen an increase in both soluble Co-60 (1.4X to 3X increase) and insoluble Co-60 (2X to 50X), resulting mainly from the release of material from fuel surfaces. Since incorporation of Co-60 on reactor surfaces usually contributes 80 to 90% of the shutdown dose rate, it may be surprising that dose rates at all NMCA plants have not increased significantly. However, testing during the qualification phase of NMCA in the mid 90's showed that noble metal treated coupons experienced greatly reduced pickup of Co-60 when exposed under simulated BWR conditions with both hydrogen and zinc additions in the water. Thus, even with the increase in reactor water Co-60 under post NMCA conditions, the addition of zinc at 5 ppb or greater can offset the tendency for increased dose rates. Those plants that had well established zinc injection programs and increased feedwater zinc to maintain 5 ppb or greater in the reactor water immediately after NMCA had unchanged or lower post NMCA dose rates.

The observed increases in reactor water Co-60 and other isotopes are similar to those that occur when a plant first initiates standard HWC, except that the effects are magnified with NMCA. When the environment is changed from oxidizing to reducing, there is a change in the stable form of oxide, resulting in a conversion of hematite (Fe_2O_3) to magnetite (Fe_3O_4). This change releases both soluble and insoluble species from fuel surfaces. With NMCA, this change occurs over all treated surfaces at the same time, creating a long-lasting increase in soluble and insoluble Co-60 and other species. As with the shutdown dose rate increase seen with HWC, zinc in the reactor water mitigates the dose rate increase following NMCA, both by suppressing the release of Co-60 from fuel deposits and by competing with cobalt for the same tetrahedral crystal sites in spinel corrosion films.

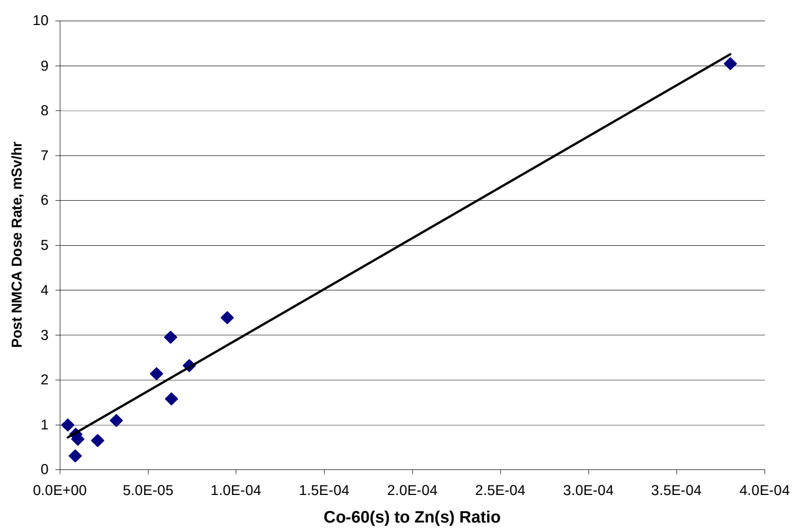

Figure 2: Measured post NMCA shut down dose rates vs. the reactor water Co-60(s) to Zn(s) ratio

Theory predicts that because zinc ions and Co-60 ions compete for the same space in the lattice of spinel corrosion films and crud, the lower the ratio of reactor water Co-60 to reactor water zinc, the less Co-60 will be incorporated into new films forming during the restructuring process that occurs in post NMCA operation. The trend line in Figure 2, which plots the post NMCA shut down dose rate versus the reactor water Co-60 to Zn ratio.

Clearly, the lower the ratio, the lower the subsequent shut down dose rate, as predicted by theory. Even though in all cases the soluble Co-60 level increased in post NMCA operation, those plants that maintained a zinc reactor concentration in the 5 to 10 ppb range saw either no change or a decrease in dose rates. In summary, the post NMCA dose rates are primarily controlled by the ratio of reactor water Co-60(s) to Zn(s). Other secondary factors, such as feedwater iron level and length of time prior to the NMCA application that HWC and feedwater zinc injection have been employed also play a role.

III. PWR ZINC INJECTION

The current trend toward longer fuel cycles in PWRs has placed an added concern on optimization of RCS chemistry. Despite application of an optimum chemistry control program, higher radiation fields may be observed. The reduced frequency of outage-related work, along with proper work planning and application of shutdown chemistry controls may decrease the impact of longer fuel cycles.

Laboratory studies, increasingly comple-mented by experience in operating PWRs, indicate a benefit of zinc additions to the reactor coolant system as a means to effect dose rate reductions and potentially mitigate the occurrence and severity of primary water stress corrosion cracking of Alloy 600.

EPRI and Southern Nuclear cosponsored the initial field demonstration of zinc addition at Farley Unit 2 in 1994-95. The results of this demonstration confirmed the beneficial effects of zinc in mitigating radiation fields, which is now well established with positive results observed in both domestic and German PWRs. A more elusive issue has been the effectiveness of zinc in mitigating PWSCC. Although laboratory testing has indicated a beneficial zinc effect on miti-gating crack initiation in Alloy 600, data for a beneficial effect on crack propagation are mixed. Accordingly, EPRI has initiated a comprehensive laboratory study to quantify the effects of zinc on primary water stress corrosion cracking (PWSCC).

Measurements of dose rates after Cycle 10 at Farley 2 with zinc addition showed a reduction of 24% at steam generator channel heads of which 11% was attributed to zinc addition and shutdown chemistry practice. Zinc-65 was less than 10% of the radioisotopic mix and only a minor contributor to the radiation fields. A dose saving of 40 man rem per fuel cycle was estimated for Farley 2 after five cycles of natural zinc addition. No zinc was added during Cycle 11 at Farley-2. Farley-2 resumed zinc addition in Cycle 12. However the period of zinc addition during Cycle 12 was too short (3 months) to assess its effect on either dose rates, fuel cladding corrosion, or PWSCC.

Diablo Canyon Unit 1 began adding natural zinc in June 1998, followed by Unit 2 in 1999. After the first application on Cycle 9, the levels of ^{58}Co in the reactor coolant increased significantly in both Unit 1 and Unit 2. Only modest activity increases were observed following Cycle 10, the second with zinc injection. The data suggest that at least one cycle with zinc chemistry is required to stabilize the zinc-substituted corrosion product deposits. Zinc injection reduced shutdown dose rates for both Diablo Canyon units. After the second cycle with zinc chemistry, steam generator dose rates were approximately 42% lower than levels prior to zinc injection at Unit 1 and 59% lower at Unit 2, although other factors may have contributed to this improvement. The activity ratios of coolant particulates for the second cycle suggest that the corrosion product deposits were stabilized through the incorporation of zinc.

Several PWRs in USA and Europe are currently injecting zinc into the primary coolant, at levels ranging from 5 to 30 ppb, with the higher levels selected to mitigate PWSCC. The data from these plants indicates that zinc addition continues to lower shutdown dose rates, but the reduction is less for each succeeding fuel cycle. This trend is reasonable, since the corrosion films are becoming conditioned with respect to exchange of nickel and cobalt for zinc. The trend seems to correlate with the cumulative exposure to zinc, as shown in Figure 3, which was developed by Westinghouse for EPRI in a published EPRI report. Plants that used depleted zinc, to avoid activation to zinc-65, showed the greatest improvement.

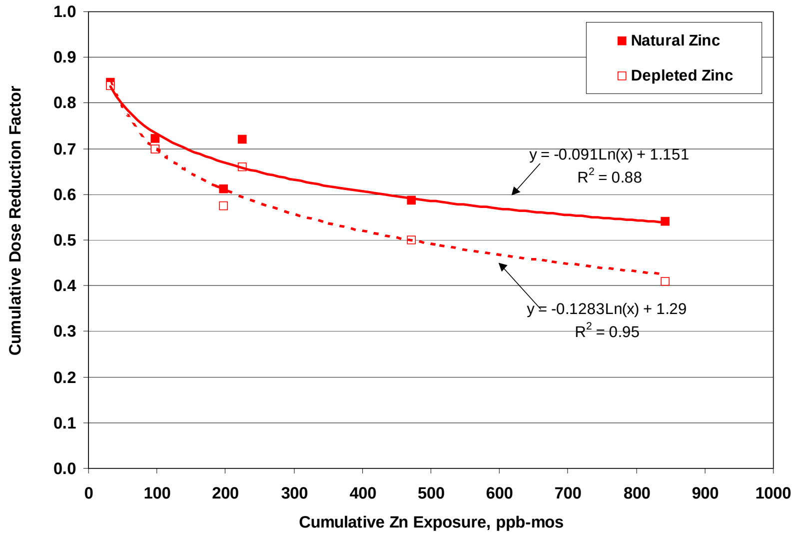

Figure 3: Cumulative dose reduction factor as a function of cumulative zinc exposure

Zinc addition to the primary coolant, even at the relatively low levels of approximately 5 ppb in the RCS, appears effective for reducing radiation dose rates, and zinc addition is being used at several plants for this purpose. Zinc is used at the 20 - 50 ppb level at Farley and Diablo Canyon to mitigate PWSCC. Consideration should be given to performance of a fuels evaluation before addition of zinc at these higher levels, particularly for higher rated cores.

The 2003 edition of the EPRI PWR Water Chemistry Guidelines recommends that each PWR consider injecting zinc. In preparing both equipment and documentation for injection at low levels for radiation control, it is prudent to plan ahead for higher injection rates if the ongoing laboratory program confirms the benefits of zinc in mitigating PWSCC.

IV. ULTRASONIC CLEANING OF NUCLEAR FUEL

When PWRs operate with higher fuel duty and longer cycles, sub-cooled nucleate boiling in the upper fuel spans is a consequence. Thermodynamic and hydraulic factors favor deposition of corrosion products on the boiling surfaces of the fuel, resulting in axially non-uniform deposition on high-duty fuel. Axially variable distribution of boron compounds in these fuel deposits is an important cause of local flux depression, termed axial offset anomaly (AOA).

Ultrasonic fuel cleaning was demonstrated to be an effective means for removing PWR fuel deposits, hence mitigating the AOA problem. In addition the reduced fuel crud inventory was shown to reduce dose rates on subsequent shutdown for refueling.

Although ultrasonic fuel cleaning has been applied at PWRs primarily for mitigation of AOA, a reduction in ex-core dose rates, and consequently personnel exposure, is also observed. These ALARA (As Low As Reasonably Achievable) benefits can be advantageously achieved at BWRs as well. The Callaway PWR has observed dose rate reductions on the order of 50% for an outage following operation with cleaned reload fuel. Such reduced radiation fields can have a significant favorable impact on personnel dose. EPRI-sponsored modeling calculations by Westinghouse for PWRs and General Electric for BWRs have confirmed that such dose reductions should be expected if the corrosion products can be effectively removed from reload fuel.

Following an extensive qualification program, fuel reliability following ultrasonic cleaning was first demonstrated at the Callaway PWR, where no fuel failures attributable to the ultrasonic process have occurred in the sixteen lead test assemblies, nor in two fuel cycles of fully cleaned reload fuel. At the time of writing, the technology has already been used subsequently at three other PWR plants in USA, and the first BWR application is planned for 2004.

Callaway has experienced AOA for many of its recent fuel cycles, and the plant staff has worked actively to mitigate this problem. The data for Cycle 12, for which all reload fuel has been cleaned, indicate that fuel cleaning is of significant value in controlling AOA. A reduc-tion in ex-core dose rates was a welcome secondary benefit. Based on the results described above, AmerenUE cleaned all reload fuel again prior to loading the core for Cycle 13 in Autumn 2002. Data on subsequent radiation fields show a significant reduction from pre-fuel cleaning (Cycle 10) to the most recent Cycle 12 (Figure 4). Although BWRs do not suffer from AOA, it is anticipated that ultrasonic fuel cleaning to remove crud from the fuel cladding surfaces will have several advantages:

1. Mitigation of potential crud-related fuel problems, especially for plants with high iron levels
2. Removal of the largest source of ^{60}Co, reducing reactor water concentrations of this isotope and resulting in lower radiation fields and reduced demand for depleted zinc
3. Fuel cleaning after NMCA will remove high concentrations of noble metal from the fuel surfaces, allowing higher loading of noble metals on core internal surfaces.

The above points suggest that ultrasonic fuel cleaning can be of real benefit for BWRs, especially if cleaning occurs immediately following NMCA application. The relatively high concentration of noble metals residing on the fuel after conventional NMCA application appears to serve as a source term for replacing the non-fuel noble metals as the latter are eroded or eluted from the plant components while in service. If such replacement redistribution from fuel to non-fuel surfaces has been a significant mechanism, the presence of noble metals on fuel may significantly increase the minimum interval between reapplications of NMCA. It may therefore be necessary to use higher noble metal concentrations initially to avoid more frequent reapplication of NMCA if the fuel is cleaned immediately following NMCA.

The option of cleaning BWR fuel was first considered (but not used) at River Bend in 1999. Mockup and laboratory tests were conducted, forming the basis of a preliminary conclusion that fuel cleaning could be an effective method for recovering highly-crudded BWR fuel assemblies.

As part of the final design qualification process, two sections of a fuel rod discharged after three cycles with NMCA were ultrasonically cleaned in the Vallecitos Nuclear Center hotcells during June 2003. Overall, no negative impact of ultrasonic energy on fuel pellets was observed.

A BWR fuel cleaner was designed in 2003; with the goal of constructing a prototype cleaner for demonstration tests in the Quad Cities fuel pool early 2004, followed by cleaning of reload fuel during the Quad Cities 2 refueling outage in February 2004.

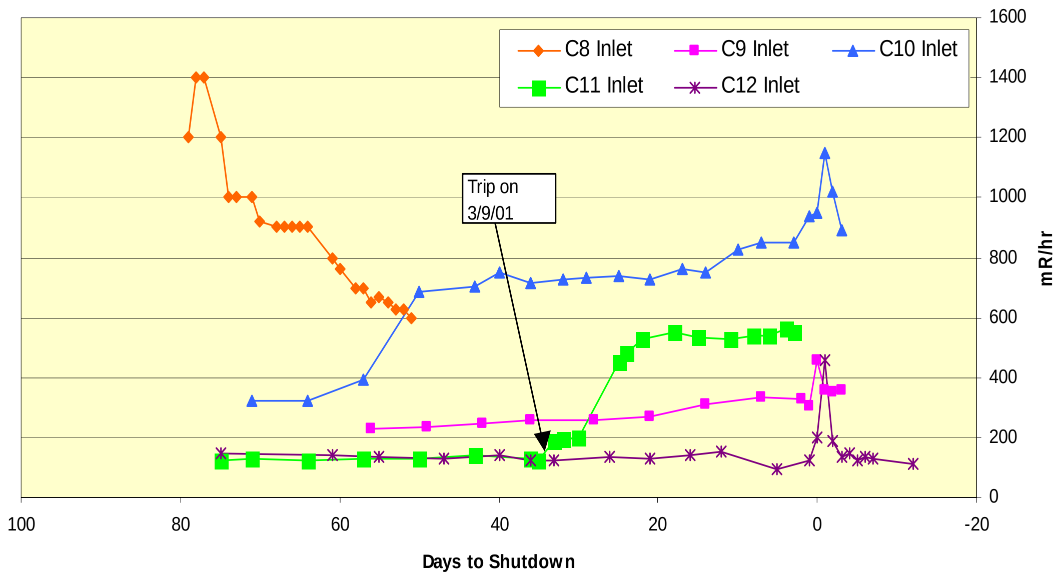

Figure 4: Callaway Letdown Heat Exchanger Inlet Dose Rates

V. RADIATION FIELD CONTROL MANUAL

The EPRI Radiation Field Control Manual has been used as a reference by RP managers, chemists, engineers and executives for many years. The manual was last updated in 1997, and several new technologies and methodologies have been developed and employed since then. A new version of the manual is to be written with few references to the previous manuals. The topics discussed in the manual will include:

- Radiation field origins and countermeasures —A review of radiation field sources, and an overview of the methods to prevent and reduce them

- Source term reduction—Discussing the development and application of hardfacing alloys (such as EPRI's NOREM) in valves and control rod blades, as well as reviewing the impact of low-cobalt steam generator tubing on fields.

- Surface preconditioning—A review of the development and effects of surface pretreatments such as electropolishing, pre-oxidation, and EPRI's Stabilized Chromium Process (SCrP). New information will include the effects of electropolished steam generator channel heads.

- Effects of PWR primary chemistry on radiation fields—A review of the various primary coolant chemistry strategies is discussed, as well as the effects of zinc addition on fields is discussed. A discussion of enriched boric acid will also be included.

- BWR coolant chemistry effects on radiation fields—A review of the interactions and results of hydrogen water chemistry, natural and depleted zinc oxide addition, and noble metals chemical application is discussed.

- Chemical decontamination—A variety of dilute chemical decontamination (DCD) techniques and their various applications will be described.

- Ultrasonic fuel cleaning—The impacts of ultrasonic fuel cleaning on ex-core surfaces, as well as an introduction to the exposure risks involved in the waste disposal, is presented.

- Appendices describing the Quad Cities and Brown's Ferry Unit 1 restart dose reduction initiatives will be added as practical demonstrations of the concepts described in the manual.

The Radiation Field Control Manual will be published and available to EPRI members in December, 2005.

VI. CONCLUSIONS

The three technologies described in this paper have been successfully introduced at operating plants. These examples demonstrate the close interaction between mitigation of materials degradation, fuel performance issues and radiation exposure concerns. It is interesting to note that these advances take advantage of synergistic benefits, producing win-win situations:

- The combination of HWC/NMCA/Zinc in BWRs mitigates stress corrosion cracking and reduces radiation fields.

- Zinc in PWRs reduces radiation fields and appears to mitigate PWSCC.

- Ultrasonic fuel cleaning addresses fuel performance issues in both PWRs and BWRs, and also reduces radiation fields.

In fact, these combinations of benefits facilitate the introduction of new technology, which is sometimes difficult to justify economically on radiation exposure grounds alone.