Corrosion Product Generation, Activity Transport and Dose Rate Mitigation in Water Cooled Nuclear Reactors

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Contents

1 Introduction

Adhering to ALARA (as low as reasonably achievable) principles is critical to achieve an effective radiation protection program in NPPs that would result in lower collective doses and lower individual doses. Activated corrosion products generated in the coolant, common to all water-cooled nuclear reactors, play a key role when they deposit on out-of-core surfaces, particularly in areas such as BWR recirculation piping, PWR channel heads and around valves, where inspection and routine maintenance work is performed.

In water cooled nuclear reactors, corrosion product generation in the coolant heat transport circuit is inevitable due to structural material interaction with the high temperature reactor coolant. The corrosion product generation is a complex multistep process that involves release of ionic species from the structural material or oxidized surfaces in contact with the high temperature reactor coolant, hydrolysis of cations leading to hydroxides, oxides, oxyhydroxides and other metastable species, generation of colloids and subsequent formation of particles of different dimensions and sizes. These initial basic physicochemical steps apply to all water-cooled nuclear reactors regardless of the reactor type BWR, PWR or VVER. However, the nature and extent of the corrosion products generated will be different, depending on the reactor type due to the different reactor structural materials used, reactor temperature, water chemistry including pH, fluid dynamics and thermal hydraulics.

Once the corrosion products are generated, they get transported to the core and undergo activation on fuel surfaces generating a variety of undesirable activated corrosion products, some of which have high gamma energies and long half lives creating an adverse radiological condition in the plant. Some of the key activated corrosion products include Co-60, Co-58, Mn-54, Cr-51, Fe-59, Zn-65, Ag-110m, Sb-122 and Sb-124. The main source of Co-60 is the stellite hardfacing alloys used in control rod drive systems, valve seats and jet pump wedges (BWRs). The main sources of Co-58 are the nickel alloys like Alloy 600 and Alloy 690 used in steam generator tubing, upper and lower head penetrations, core support and pressurizer nozzles (PWRs), and transition attachment materials used between stainless steel and reactor pressure vessel material, shroud head bolts, access hole covers and shroud support (BWRs). The main source of antimony activation products are the bearing materials and main pump seals impregnated with antimony (PWRs and VVERs).

Once the activated products are generated, they get transported throughout the heat transport circuit depositing on out of core surfaces, by incorporating into the oxide film as ionic species or by depositing on the oxide film as particulates, creating undesirable dose rates and "hot spots". It is believed that the generation of radiation fields in the high flow areas of a BWR like the recirculation piping is largely controlled by the incorporation of soluble activated species like soluble Co-60 into the oxide film. On the other hand, in the low flow areas, the dose rate generation may be largely controlled by particulate deposition leading to "hot spots". The contributions arising from each of the activation products to plant radiological conditions are different for different reactor types due to the usage of different structural materials, water chemistries, fluid dynamics and temperatures.

However, the basic processes involved in enhanced radiological conditions in a power plant can be summarized as follows:

- Generation of corrosion products due to corrosion of structural materials in contact with the reactor coolant

- Release of corrosion products in to the coolant

- Transport of corrosion products in the heat transport circuit

- Deposition of corrosion products on fuel surfaces

- Activation of elemental species to activated corrosion products on fuel surfaces

- Release of activated corrosion products from fuel surfaces into the coolant

- Transport of activated corrosion products on to out of core surfaces

- Incorporation or deposition of activated corrosion products on out of core surfaces

- Generation of increased radiation fields in the plant during operation, start-up and shutdown

This report discusses in detail, the steps involved in, generation of corrosion products including colloid formation, activation on fuel, transport through the coolant, deposition on surfaces including zeta potential effects, release from surfaces and removal of activated corrosion products in light water reactors. The report also discusses activity transport that will include basic steps involved and models used. The report is largely devoted to BWR experiences, however some limited examples of PWR and VVER experiences are also included for the benefit of non-BWR scientists, since the basics and physical principles involved in dose rate generation and management have many similarities to all reactor types.

A discussion of dose rate mitigation technologies used to control radiological consequences to nuclear power plants including,

- Material replacements

- Water chemistry controls/modifications

- Chemical additives

- Surface treatments

- Cobalt filtration

- Fuel cleaning

- Decontamination approaches

- Plant operational strategies

- Occupational exposure

are also included in the report. This report is expected to be a comprehensive summary of radiation field generation and mitigation approaches that will be of benefit to nuclear power plant personnel.

The report does not address the effect of detailed operational history, core design, fuel design and fuel failures and their impact on the radiological conditions of nuclear power plants.

2 Corrosion Product Generation in NPPs

Structural materials like stainless steels (Type 304, 304L, 316, 316L, 347, 321) and nickel base alloys (Alloy 600, 690, 800) utilized in NPPs react with the high temperature coolant during plant operation forming oxide films on the surface. Oxide layers are formed as duplex layers with a relatively thin compact inner layer adjacent to the metal. The outer oxide tends to be more porous formed largerly by a dissolution precipitation type mechanism, allowing incorporation of anions and cations from the coolant into the oxide layer.

The inner layer tends to be in a more reduced state owing to the less accessibility to oxygen and oxidants, while the outer layer is in an oxidized state because of the ready accessibility to oxygen and oxidants in the coolant. Therefore, in the case of iron base alloys the inner layer tends to be the more reduced magnetite (Fe_3O_4) form, while the outer oxide tends to be of hematite (Fe_2O_3) form. Following a similar approach, the inner oxide on stainless steels tends to be the reduced form of non-stoichiometric Ni, Fe, Cr oxide ($Ni_xFe_{1-x}Cr_2O_4$) or the stoichiometric nickel chromite ($NiCr_2O_4$). The outer oxide tends to be the oxidized form of non-stoiciometric Ni, Fe oxide ($Ni_xFe_{1-x}Fe_2O_4$) or the stoichiometric nickel ferrite ($NiFe_2O_4$) as shown in Figure 2-1.
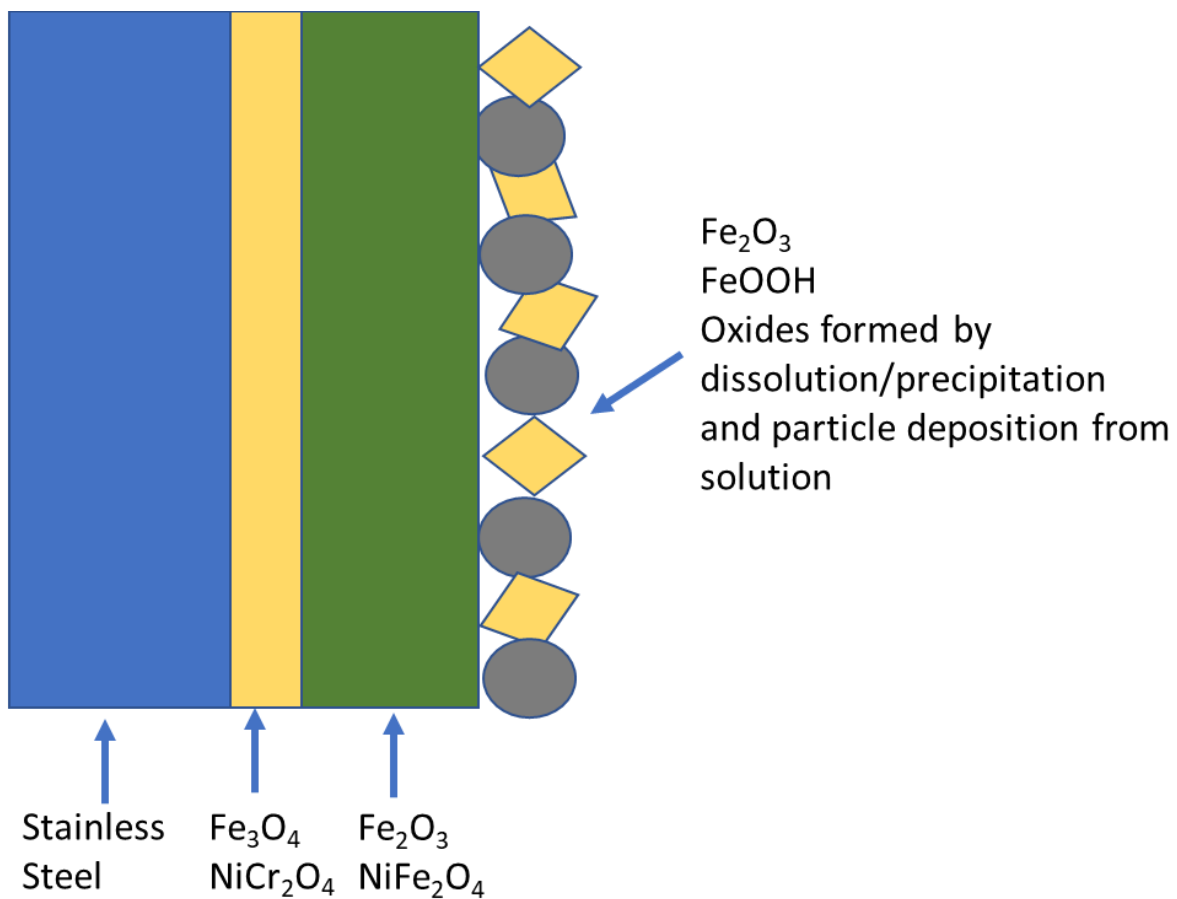
Figure 2-1: A schematic of inner and outer oxides formed on stainless steel along with outer surface layer formed by dissolution/precipitation and deposition of particluates from the solution side.

3 Cobalt-60 Generation on Fuel Deposits and their Composition

If elemental cobalt (^{59}Co) that enters the reactor water from the sources shown in Table 2-1 were to stay in the water, very little ^{60}Co would be produced and ^{60}Co would not be a major contributor to shut down dose rates. Unfortunately, soluble ^{59}Co reacts with insoluble iron oxide particles that reside on fuel rods deposited as crud. The deposition mechanism of soluble and insoluble impurities into a solid phase on fuel deposit is shown in Figure 3-1. Figure 3-1 (a) shows the growth of steam bubbles trapping and depositing particles from the reactor water on to the surface of the fuel cladding. Figure 3-1 (b) illustrates the thickening of the crud layer by wick boiling, and Figure 3-1 (c) depicts the mature fuel deposit with the underlying tenacious layer covered by the loose layer on top. The tenacious layer is formed by a hydrothermal sintering type process which is likely accelerated by the effects of high neutron flux. The loose layer is called the "brush" layer, because it is readily removed during crud sampling by using a soft bristled brush. The tenacious layer is called the "scrape" layer because it can only be collected during sampling by scraping the surface with a grinding stone. Over 99% of the ^{60}Co inventory resides in the "brush" and "scrape" layers of fuel deposits.
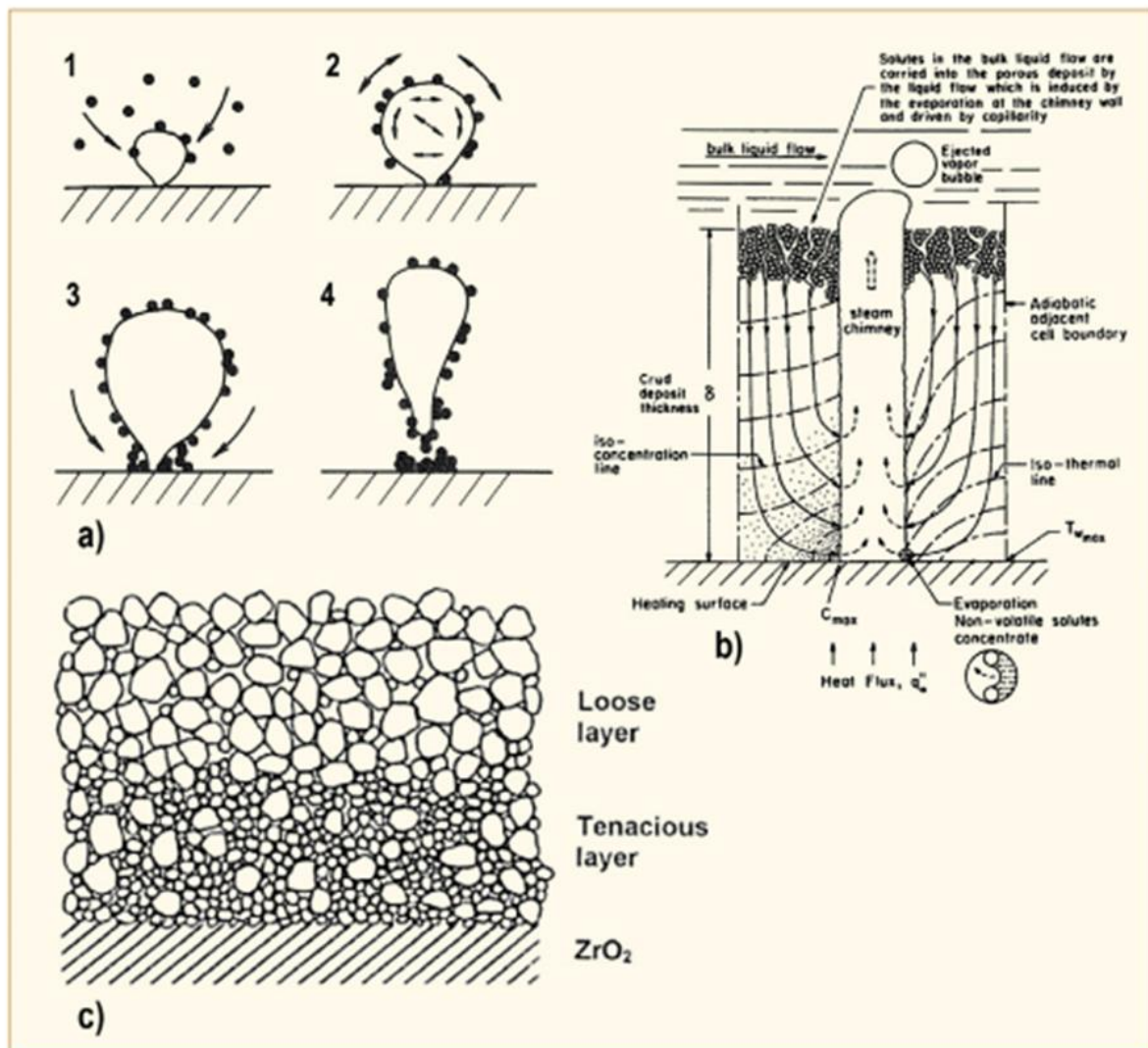
Figure 3-1: Schematic diagram showing, a) bubble formation along with particle agglomeration, b) wick boiling with steam chimney, and c) a mature fuel deposit [Cowan et al, 2012].

4 Incorporation of ^{60}Co into Oxide Films

Laboratory data showed that the Co/Fe ratio in the corrosion film is proportional to the soluble Co concentration over the range typical of that found in BWRs [Niedrach, 1980]. Note that 0.1 µg /l of Co in the water phase resulted in a 10^8 concentration factor of Co in the stainless steel oxide. When reactor water soluble ^{60}Co was plotted against the *BRAC dose rates for newer plants or had new recirculation piping from both US and Japanese BWRs that were either new plants or had new recirculation system piping a linear relationship was seen as shown in Figure 4-1. Only plants operating with normal water chemistry (NWC), with or without Zn injection, were included in this plot [Anstine 1983, Lin 1995].
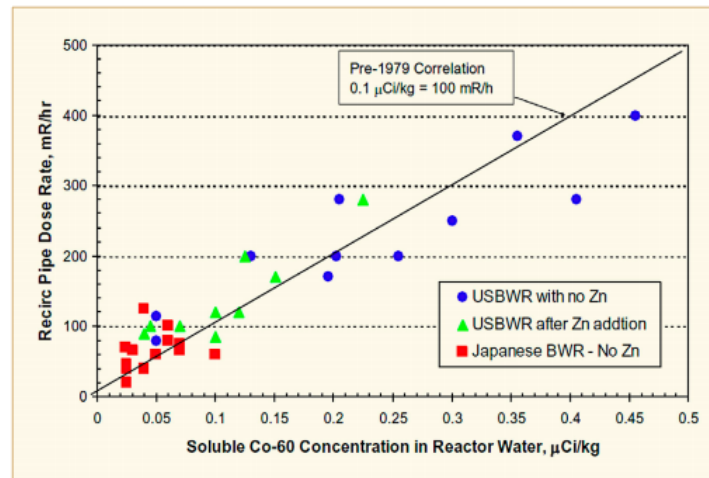
Figure 4-1: Correlation of recirculation piping dose rate with ^{60}Co(s) in reactor water under NWC conditions (pre-1985) [Lin, 1995].

When the values of deposited ^{60}Co are measured on recirculation piping at reactor outages and plotted against the measured BRAC values, Figure 4-2, the results are linear with an exceptionally good correlation factor of 0.94. The ^{60}Co concentrations on the piping have been measured with a calibrated, heavily shielded gamma detector. In all cases the gamma radiation from ^{60}Co dominates the BRAC value, whether the environment was oxidizing or reducing or in the presence or absence of Zn.
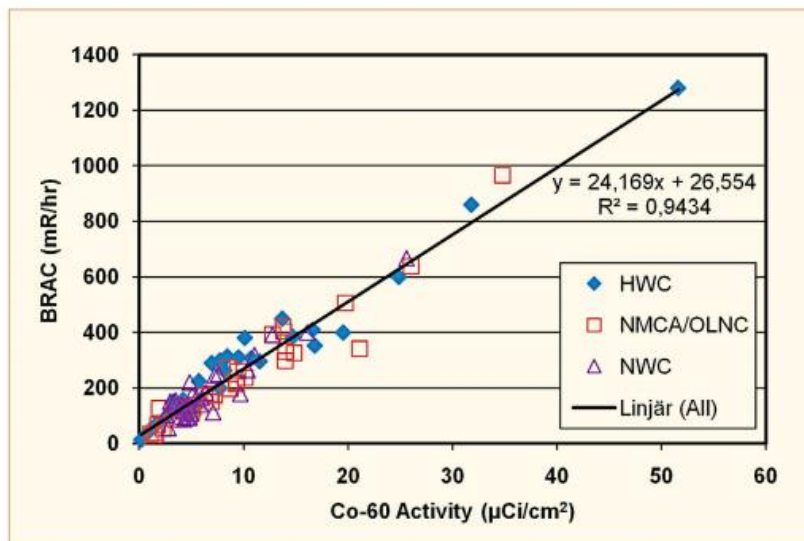
Figure 4-2: Correlation of ^{60}Co measured on recirculation piping surfaces during outages as a function of the measured BRAC values [Jarvis & Wells, 2011].

5 Activity Transport and Modeling

The basic mechanisms involved in generation, activation and transport of radionuclides consist of many steps that are generally independent of the type and water chemistry of the NPPs. The specific details of the type radionuclides formed, their concentrations, transport rates, deposition rates, release rates and the generated dose rates may vary depending on the NPP type.

5.1 GE Cobalt Transport and Shutdown Drywell Dose Rate Model for BWRs

The basic processes involved can be summarised as follows [Lin, 1993]:

- Corrosion product generation from primary system surfaces or entry through feedwater. Corrosion products are in ionic form, colloidal form or in larger particulate form

- Transport of corrosion products on to fuel

- Depostion of corrosion products on fuel fuel surfaces by van der Waals or electrostatic forces. Deposition may be promoted by heat flux and bubble evaporation

- Activation of corrosion products on fuel by the neutron flux. Their residence time may vary from days to weeks. Table 5-1 shows a list of most common radionulcides generated their half lives, gamma enrgies and the formation reactions

- Release of radionuclides from fuel in the form of ions, colloids or particles. Release may occur by dissolution, flow induced hydraulic shear, erosion and particle spallation

- Transport of radionuclides on to out of core surfaces

- Incorporation of ionic forms such as ^{60}Co into the oxide films such as piping generating out of core radiation fields. Deposition of particulates on out of core surfaces by van der Waals or electrostatic forces creating hot spots in the pedestal area or control rod drive housings

- Release of some radionuclides by crud release into the coolant due to solubility differences during shutdowns and outages

- Removal of the activation products by the clean up system that maintains a steady state concentration of the activation products

In terms of total ^{60}Co inventory, over 99% remains locked in the fuel deposits and less than 1% is deposited in out of core locations. However, because of the high gamma energies of ^{60}Co, the 1% in out of core locations can cause significant operational issues and exposures. Figure 5-1 shows a flow diagram that can form the basis for computer models to make calculations on a plant specific basis that predict reactor water ^{60}Co concentrations and shutdown dose rates [Lin & Garcia, 1994].

A GE dose rate model based on the flow path shown in Figure 5-1 was successfully used to predict shut down dose rates of BWRs with and without zinc addition. The model was able to predict shutdown dose rates of several BWRs at different levels of zinc addition, proving the predictive capability of the model. This is the most comprehensive BWR dose rate model that has been used widely in GE BWRs.

There are few other BWR models like the Tsuruga model (Japan), Hitachi DR CRUD model (Japan), Toshiba BWR Cobalt Transport Model, and ASEA-Atom BWR crud model (Sweden) that have been used to understand and predict dose rates in BWRs, with some variations [Alder et al, 1992a&b], [Gregorich, 2016].

6 Cobalt Source Term Reduction

A wide range of reactor water ^{60}Co values has been reported in operating BWRs, with cycle median values ranging from 0.5 to 16 Bq/ml, as shown in Figure 6-1. This figure shows data from GE BWR fleet in the US, Mexico and Taiwan. Under steady state conditions, regardless of the oxidizing conditions, the age of the oxides on the stainless surfaces or the type of FW additives, a reduction in the reactor water ^{60}Co(s) concentration should result in an eventual reduction in the steady state concentration of ^{60}Co incorporating into the oxide film. Most BWR operators include the reduction of reactor water ^{60}Co in their strategic plans for the future improvement of their plants. This section discusses the strategies used to achieve source term term reduction in NPPs.
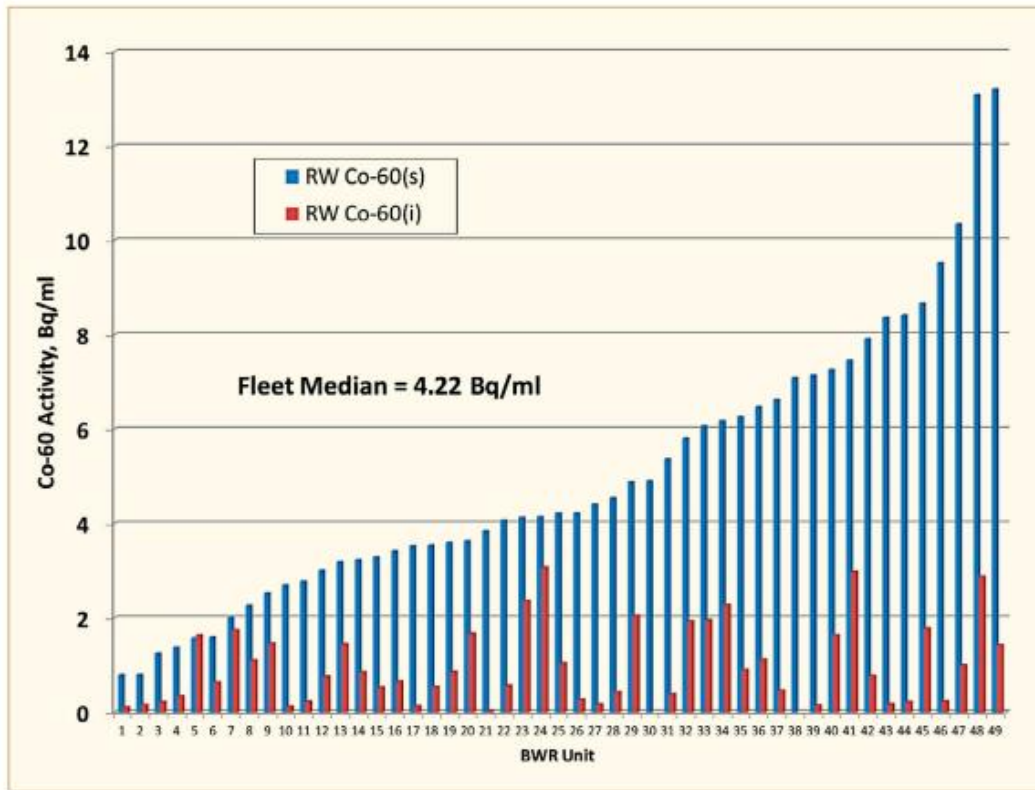
Figure 6-1: Median cycle reactor water ^{60}Co(s) for the GE BWR fleet as of 2011 adapted from [Cowan et al, 2012a].

6.1 Cobalt Source Term Reduction Strategies

The naturally occurring cobalt isotope is ^{59}Co, which gets activated via an (n,γ) reaction to ^{60}Co. Thus, if natural cobalt isotope can be prevented from activating in the core of the reactor, then ^{60}Co generation is eliminated. There are three main ways to limit the natural ^{59}Co sources as follows:

- Replace out of core cobalt containing components that can corrode or wear and act as a cobalt source term via the feed water or by corrosion of the systems that recirculate through the reactor water

- Improve filtration and ionic removal of elemental cobalt in water streams

- Replace cobalt alloy containing components in-vessel near the core region that can be activated in situ and then release ^{60}Co into the reactor water

7   Zinc Addition for BWR Dose Rate Reduction

It was observed that BWRs with soluble Zn ion (arising from admiralty brass condenser tubing) in the reactor water tended to have significantly lower shutdown dose rates. A hypothesis was developed that zinc cations incorporate into and modify the normal magnetite crystal defect structure so that a more protective film is formed and corrosion is significantly reduced. It was found that BWRs with significant Zn content in the corrosion film had lower BRAC dose rates [Marble & Wood, 1985].

GE and EPRI launched a laboratory program consisting of two parts, a) confirmation of the incorporation of Zn ion from solution into stainless steel corrosion films and b) the effect of zinc ion in solution on the uptake of 60Co into the corrosion film. The laboratory results showed that the zinc in the water made stainless steel oxide films thinner and the incorporation of 60Co into the oxide film was significantly lower. Furthermore, it was confirmed the zinc ions need to be present in the water in order to lower the cobalt uptake by the corrosion film.

Hope Creek was the first BWR to implement zinc addition in to the feedwater. Hope Creek is a BWR-4 with an original power rating of 1117 MWe with a titanium condenser and a Deep Bed (DB) cleanup system. The initial criticality was in June of 1986 and zinc injection began in January of 1987 when the final feed water reached a high enough temperature to assure complete dissolution of the zinc oxide particles (176°C). The zinc oxide was injected into the feed water with a metering pump injecting an agitated dilute slurry of natural ZnO. The particles dissolve in seconds at BWR final feed water temperatures. The objective was to establish a reactor water soluble zinc (Zn(s)) concentration of 10 to 15 µg/kg during the first 2000 hours, the "conditioning period", and then decrease the concentration to the "maintenance range" of 5 to 10 µg/kg. A schematic of the injection system is shown in Figure 7-1. In general, the conditioning period objectives were met but the maintenance concentrations were on the low side or slightly below the target range. Hope Creek had a fairly high feed water iron input (cycle average of about 8 µg/kg) and the iron colloid particles adsorbed the zinc ions, resulting in lower than desired soluble zinc, even at the highest feed water addition rate. This was especially true during the second half of the first cycle when the feed water iron increased significantly.
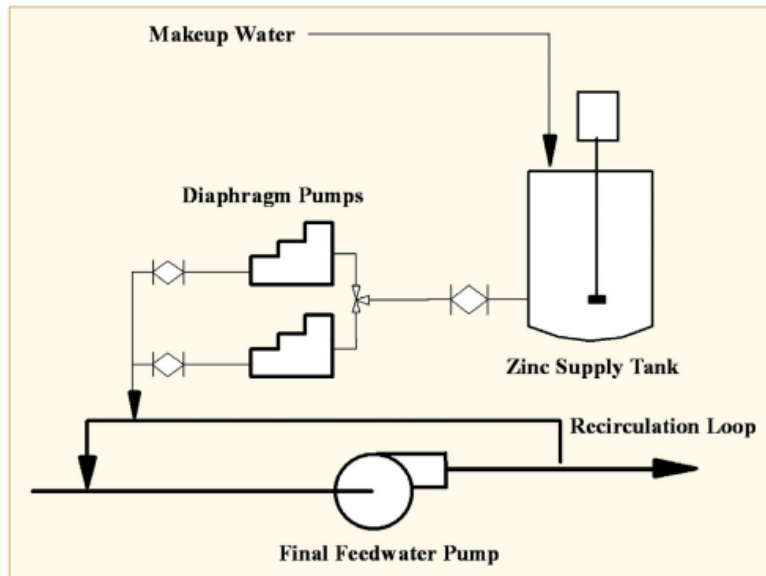
Figure 7-1:   Typical active ZnO injection system [Marble, 1994].

The BRAC dose rates were a factor of 2 to 3 lower than the industry average. However, during the first RFO at Hope Creek in the spring of 1988, there was a significant release of ^{65}Zn from the fuel deposits to the reactor water. The impact of the release was magnified significantly by the unavailability of the Reactor Water Cleanup System (RWCU). As a result, the outage activities were adversely affected.

8   Surface Treatments for Dose Rate Control

A variety of surface treatment methods have been used by plant owners to mitigate ^{60}Co incorporation into oxide films. Some of these methods are in-situ and some are ex-situ. Furthermore, some can be applied following decontaminations and some for new components used as replacement parts.

8.1   Hi-F Coat Surface Treatment Process

Hi-F Coat is a process developed by Hitachi-GE to form a fine oxide film (Magnetite: Fe_3O_4) on the decontaminated surface of piping and equipment. The fine oxide film prevents the cobalt accumulation into the film. To enhance the effects of Hi-F Coat, Shimane Unit 1 applied NWC (Normal Water Chemistry) pre-oxidation operation. NWC pre-oxidation operation is a method to stabilize the Hi-F Coat film under NWC conditions by stopping the hydrogen injection during the plant start-up period. As a result of this countermeasure, cobalt accumulation in the primary recirculation (PLR) piping was suppressed and dose rate of the PLR piping of Shimane Unit 1 was decreased [Uemura et al., 2014].

The application procedure of the Hi-F Coat is shown in Figure 8-1. The Hi-F Coat is an artificial fine oxide film made of magnetite (Fe_3O_4) and is formed on the surface of piping and equipment by injection of iron formate ($Fe(HCOO)_2$). The iron formate is injected after finishing the chemical decontamination step by using the same injection systems used in the decontamination process. The process uses three chemical reagents: iron formate, H_2O_2 and hydrazine. Planned values of iron and hydrazine are 250 ± 50 ppm and from 200 to 600 ppm, respectively. The process is performed with the same equipment as the HP CORD UV decontamination process and at the same temperature. Because laboratory tests showed that better results were observed on Hi-F Coat treated surfaces if they were exposed to NWC conditions for 50 days before HWC conditions were established, the results of this dual NWC/HWC exposure are shown in Figure 8-2. The mechanistic steps involved in dose reduction by the Hi-F coating process are shown in Figure 8-3.

It is estimated that the application of Hi-F coat process at Shimane Unit 1 reduced the dose rates of primary recirculation piping (PLR) piping by 50%. The process was later applied to Shimane Unit 2 as well.
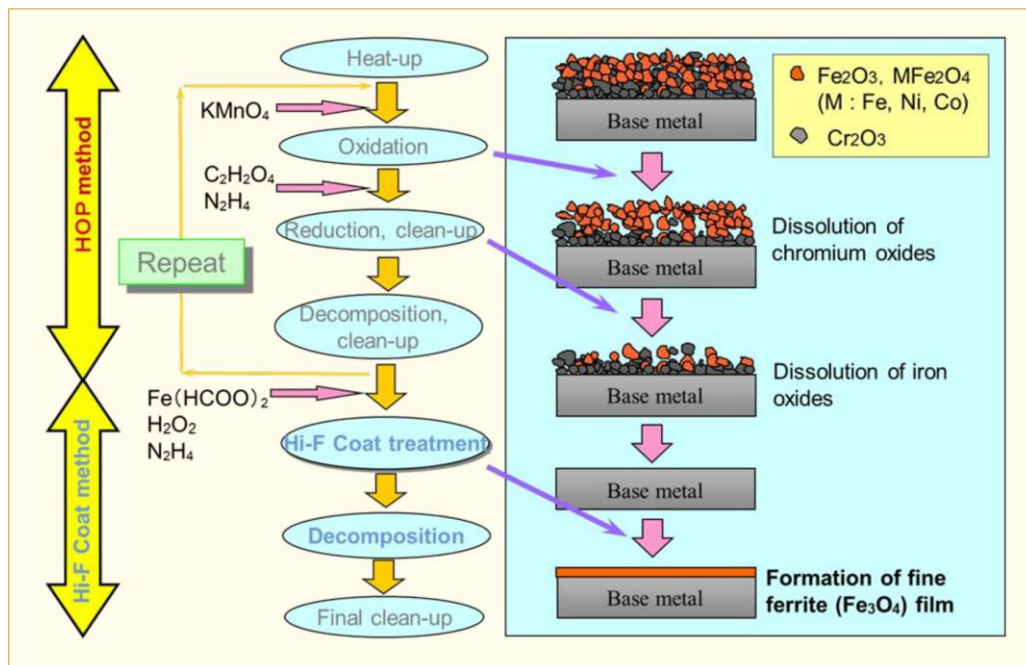

Figure 8-1:   Hi-F coat process steps involved in the application [Uemura et al., 2014].

9   Plant Operational Stratagies to Control Dose Rates

As a consequence of BRAC dose rate increases in BWRs, plant owner's attention turned to dose accumulated by personnel during plant outages in order to control occupational exposure. The focus was to explore plant operational strategies that could potentially lessen the release of activation corrosion products from fuel deposits, by retaining them on fuel deposits to the extent practical. Since the primary source of ^{60}Co, ^{58}Co and other activated species is the fuel deposits, any changes in properties that could change the kinetics, magnitude and timing of the releases by operational means is worth exploring. As an example, the change from reducing conditions with HWC to oxidizing conditions after H$_2$ injection is stopped can cause increased concentration of both soluble and insoluble corrosion products. During cool down, the solubility of cobalt, nickel and zinc spinel phases increases. Thus, it is imperative that reactor operator must plan for these changes and explore possibilities to minimize their adverse effects with any available operational changes. This section of the report deals with NPPs that have adopted measures to control the release of radionuclides from fuel deposits by a variety of operational changes.

9.1   US Approach

EPRI has issued a Shutdown and Start-up Chemistry Experience Sourcebook [EPRI, 2009] that has invaluable insights and experiences for mitigating activity release during shutdowns. The key strategies are as follows:

- Maximize cleanup systems availability, including RWCU, fuel pool, submersible filters and demineralisers.

- If the cleanup systems must be removed from service for maintenance, schedule the maintenance for a time period for after fuel moves have begun.

- Temporary cleanup systems should be available and ready to use.

- Establish performance criteria for cleanup systems.

- Have sequence specific ^{60}Co goals and strategies.

An example of a plant specific plan for ^{60}Co is shown in Table 9-1. An example of a plant shutdown ^{60}Co trend is shown in Figure 9-1. The evolution stages are: shutdown/scram, shutdown cooling start, reactor floodup, cavity floodup, gates open and RWCU out of service. The high source term plants have the highest maximum ^{60}Co(t) values for each sequential evolution stage and the low source term plants the lowest. When both source term and feed water iron are considered, the low source term/low iron combination had the lowest ^{60}Co(t) releases.

10 NPP Decontamination for Dose Rate Control

With BWR plant operation over time, ionic species and particulate corrosion products accumulate on heat transfer surfaces such as nuclear fuel. Due to its slow corrsion in BWR water, stainless steel produces a variety ionic species like elemental iron, nickel, chromium, manganese, cobalt etc. When these species deposit on fuel, they get activated some of which produce long half life gamma emitting isotopes like ^{58}Co and ^{60}Co. These isotopes get transferred to out of core surfaces creating dose rates where it is not needed. Therefore, these surfaces need periodic decontamination to remove undesirable activation products in order reduce personnel occupational exposure.

10.1 BWR Decontamination Experiences in Europe and the USA

The introduction and adoption of hydrogen water chemistry into BWRs in the US, Europe and Mexico to mitigate IGSCC concerns led to an increase in shutdown dose rates at many of the plants. Although feed water depleted Zn injection was eventually initiated at most BWRs to combat the increased dose rates, many were still dealing with fairly high dose rates due to the presence of other species such as copper in the reactor water. Copper incorporation in the oxide film appears retain more activated corrosion prodcts like ^{60}Co and ^{58}Co in the films. When the dose rates reach high values that can affect outage work due to high personnel exposure, it is imperative that these surfaces need to be decontaminated to remove the activation corrosion products. As an example, Figure 10-1 shows the number of recirculation system decontaminations since 2000.

Because of the use of DZO injection, HWC and NMCA/OLNC technologies in a large majority of BWRs in the US and some in Europe and Mexico, the oxide films formed on the stainless steel surfaces of these plants are primarily of mixed metal chromites ($MeCr_2O_4$) and metal ferrites ($MeFe_2O_4$) where the "Me" is Zn, Ni and Fe in various proportions depending on whether the oxide is stoichiometric or non-stoichiometric. Some of these oxides like the chromites are hard to remove from surfaces, and hence the decontamination formulations need to contain more aggressive chemicals. Therefore, the decontamination systems and formulations have been modified to deal with these hard to remove adherent oxides.
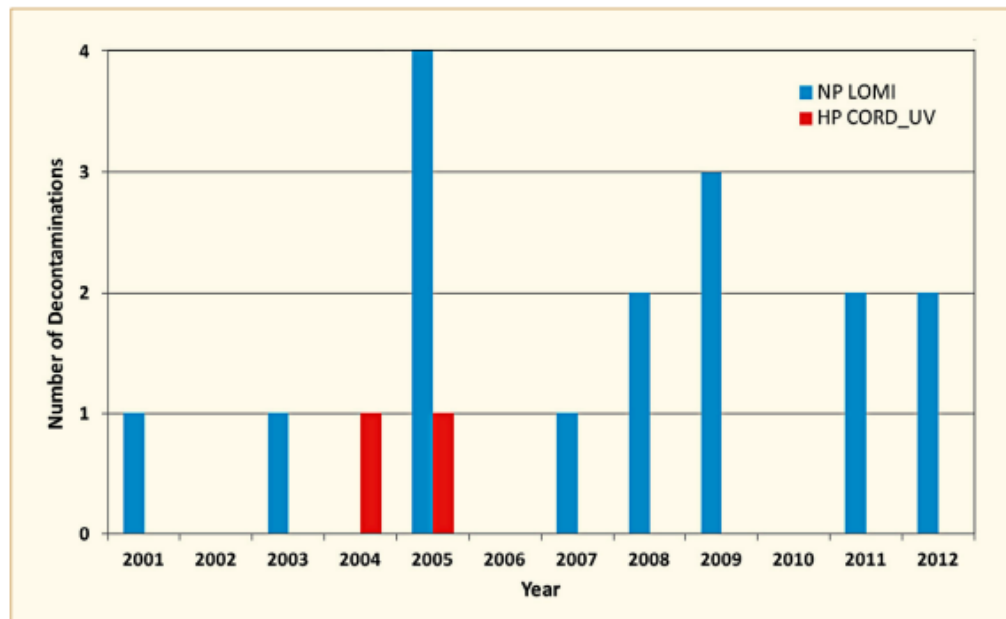

Figure 10-1: Summary of recirculation system decontaminations for American and European GE BWR fleet adopted from [EPRI, 2012].

11 Recent NPP Dose Reduction Attempts and Related Plant Experiences

This section deals with a variety of measures adopted by modern day NPPs to reduce dose in different locations of the plant to minimize personal exposure. They include, opitmization of shutdown practices to minimize crud mobilization, zinc addition to lower dose rates, cobalt control measures and plant modernizations. A few specific examples on how dose reductions have been achieved are described in this section.

11.1 Chemsitry Measures for Dose Reduction at Gundremmingen BWR

In recent years, the two Gundremmingen units experienced elevated radiation levels during outages with increased man-doses. One main radiation source was identified as being due to particle mobilization from fuel surfaces (CRUD) during shut-down operation. The temperature gradient during cool down of the reactors to refueling outages was decreased from 30K/h to 10K/h in order to reduce CRUD mobilization and to clean up the reactor water thoroughly prior to water level increase and flood up for outage work. In addition, it was decided to establish a low level of zinc in reactor water with the aim to bind activation products on the fuel rods [Nowotka et al, 2018].

The operational experience with the two units in the past with brass condenser tubes was positive. With the condenser tube material change from brass to stainless steel, the zinc effect vanished and the fuel CRUD became more mobile. It was therefore decided to inject small quantities of zinc (as zinc acetate solution) into feedwater for compensation of the loss of the natural zinc source after modernization of the turbine condensers. However, this has to be accomplished by excluding any potential formation of thermally insulating Zn-silicate layers on fuel rod surfaces [Nowotka et al, 2018].

In the past, silicate levels in the two reactors were always by a factor of 2-3 higher than in other European BWRs. Therefore, it was obvious that the silicate level had to be reduced before starting zinc injection. Reduction of silica levels was achieved by three measures:

- Identification and elimination of silicate sources in the plant filter material

- Lower water purification flow rate, along with a decrease in temperature in the precoat filters by approx. 7°C, greatly improving silicate retention

- More frequent backwashes of the powdered ion exchange resin coats of the filter demineralizers in the reactor water clean-up system, approximately ten times per 11-month cycle.

These measures proved effective and resulted in a decrease in reactor water silicate concentrations by a factor of two as shown in Figure 11-1.

Reactor Water Silica / ppb	2002/ 2003	2003/ 2004	2004/ 2005	2005/ 2006	2006/ 2007	2007/ 2008	2008/ 2009	2009/ 2010	2010/ 2011	2011/ 2012	2012/ 2013	2013/ 2014	2014/ 2015	2015/ 2016
Unit B	600	640	560	600	670	590	450	680	690	570	530	400	450	340
Unit C	-	-	620	680	680	480	550	720	510	390	330	720	540	350

Figure 11-1: Reactor water silicate cycle mean values in µg/kg (ppb), as measured in Gundremmingen units B and C between 2002 and 2016 [Nowotka et al., 2018].

The following measures were implemented to improve the power station's radiologic situation while excluding any risk for the fuel assemblies:

12 Recent Occupational Exposure Status of NPPs

Occupational exposure is a parameter that many nuclear plant operators pay attention to in order to lower the personnel exposure during a refueling outage. The whole-body radiation levels accumulated by personnel is directly related to the radiation levels produced by an NPP in and out of core surfaces. The plant radiation levels are manageable in many cases by employing a variety of new technologies available with a proactive radiation protection program.

Figure 12-1 shows the three-year rolling average collective dose per reactor for all operating reactors included in information system occupational exposure (ISOE) by reactor type from 1992 to 2017 as person·Sv/reactor.

Note: PWR is pressurised water reactor; VVER is vodo-vodyanoy energy reactor; BWR is boiling water reactor; PHWR is pressurised heavy water reactor; GCR is gas-cooled reactor; LWGR is light water graphite reactor
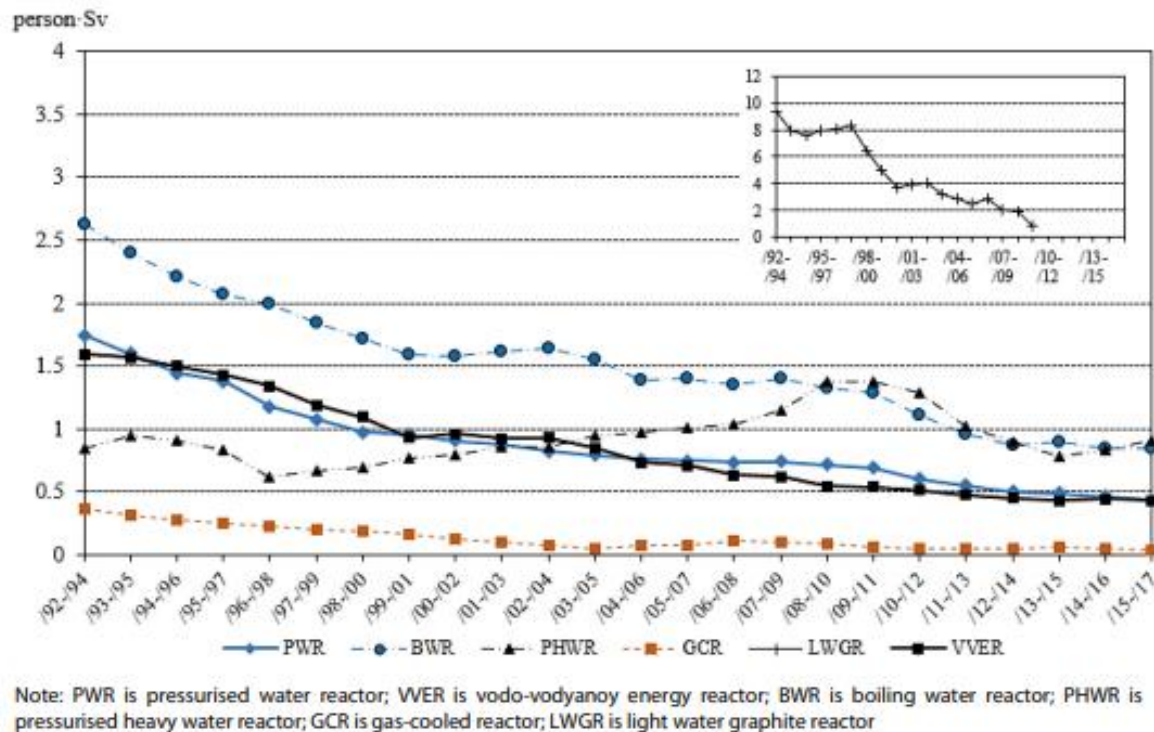
Figure 12-1: Three-year rolling average collective dose per reactor for all operating reactor types from 1992 to 2017 [NEA/OECD, 2017].

Figure 12-2 to Figure 12-4 provide information on average collective dose per reactor by country for PWR, VVER and BWR reactors. In all figures, the "number of units" refers to the number of reactor units for which data has been reported for 2017.

Clearly, PWRs have the lowest collective dose and BWRs have the highest except for those BWRs in Germany, Sweden, Finald and Japan.

13   Summary

This report provides details of corrosion product generation, their activation on nuclear fuel, transport through the heat transport circuit and deposition on out of core surfaces creating a radiologically adverse condition to the NPP. The report includes information on how the corrosion of plant materials generate ionic corrosion products which then create colloids that gets activated and deposits on surfaces depending on their surface charges and the electrokinetic properties of the colloid. If the colloidal particles are radioactive, they can get incorporated into the oxide film increasing radiation levels of the component or piping. If the coolant flow conditions are low, colloidal particles can deposit on low flow surfaces creating radiation hot spots.

The report discusses the following items:

- Generation of corrosion products due to corrosion of structural materials in contact with the reactor coolant

- Release of corrosion products in to the coolant

- Transport of corrosion products in the heat transport circuit and modeling

- Deposition of corrosion products on fuel surfaces

- Activation of elemental species to activated corrosion products on fuel surfaces

- Release of activated corrosion products from fuel surfaces into the coolant

- Transport of activated corrosion products on to out of core surfaces

- Incorporation or deposition of activated corrosion products on out of core surfaces

- Generation of increased radiation fields in the plant during operation, start-up and shutdown

This report also discusses in detail, the steps involved in, generation of corrosion products including colloid formation, activation on fuel, transport through the coolant, deposition on surfaces including zeta potential effects, release from surfaces and removal of activated corrosion products in light water reactors. The report also discusses activity transport that will include basic steps involved and models used. The report is largely devoted to BWR experiences, however some limited examples of PWR and VVER experiences are also included for the benefit of non-BWR scientists/engineers, since the basics and physical principles involved in dose rate generation and management have many similarities to all reactor types.

A discussion of dose rate mitigation technologies used to control radiological consequences to nuclear power plants including,

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- Occupational exposure

References

Alder H. P et al, "Corrosion products, activity transport and deposition in boiling water reactor recirculation systems", IAEA TECDOC Vol 3, Activity Transport Mechanisms in Water Cooled Reactors, Vienna, Austria, October, 1992a.

Alder H. P. and Brélaz P., Overview of activities for the reduction of dose rates in Swiss boiling water reactors, Water chemistry in nuclear Reactor Systems 6, BNES, London, 1992b.

Anstine L. D., BWR Decontamination and Corrosion Product Characterization, General Electric Report NEDE-12665, 1977.

Anstine L. D., BWR Radiation Assessment and control Program: Assessment and control of BWR Radiation fields, EPRI Report NP-3114, May 1983.

Asakura Y. et al., Current Operating Experience with Water Chemistry in Crud Concentration Suppressed Boiling Water Reactors, International Conference on Water Chemistry of Nuclear Reactor Systems -5, Bournemouth, UK, Oct. 23-27, 1989.

Baston V. F., Garbauskas M. F. and Oken H., Material Characterization of films on BWR Components Exposed to Hydrogen Water Chemistry and Zinc Injection, International Conference on Water Chemistry of Nuclear Reactor Systems -7, Bournemouth, UK, October 1996.

Beal S. K, Deposition of Particles in Turbulent Flow on Channel or Pipe Wall, Nuclear Science Engineering, Vol 40, Page 1, 1970.

Bockris J. O' M, Devanathan M. A.V, and Müller K, Proc. Royal Society (London) A 274, 1963.

Bridle D.A and Cake P, Water Chemistry of Nuclear Reactor Systems, Vol 2, BNES, Bournemouth, UK, October, 1992.

Brunk J., System decontamination of two BWR units performed during 2011 and 2012, ISOE 2012, Prague, 2012.

Brunning P., Bengtsson B., Granath G. and Kvint J., Corrosion Product Measurements at Ringhals 1, International Conference on Water Chemistry of Nuclear Reactor Systems -7, Bournemouth, UK, October 1996.

Burrill K. A, Nuclear Technology, 36, 85, 1977.

Burril K.A, Improving Activity Transport Models for Water-Cooled Nuclear Power Reactors, AECL Report 12076, Chalk River, Ontario, Canada, August 2001.

Chen J. et al, On Cobalt Deposition in Oxide Films formed on Stainless Steel under Simulated BWR Coolant Conditions, Nuclear Plant Chemistry Conference, NPC 2018, San Francisco, CA, 10-14 September 2018.

Cowan R. L. et al, Hydrogen Water chemistry Operating Experience, Post-SMIRT Conference, Ispra, Italy, August 1985.

Cowan R. L., Lin C. C., Marble W. J. and Skarpelos, J. M., Recent Developments in BWR Water Chemistry, 2nd International Topical Meeting on Nuclear Power Plant Thermal Hydraulics and Operations, Tokyo, Japan, April 15-18, 1986.

Cowan R. L. and Marble W. J., Hydrogen Water Chemistry Effects on BWR Radiation Buildup - Preliminary Evaluation of Plant Data, EPRI TR-101463, November, 1992.

Cowan R. L., Adamson R. B., Garcia S. E., Potts G. A., Levin H. A. and Armijo J. S., BWR Water Chemistry Strategies for Simultaneously Minimizing Fuel Clad Corrosion, Mitigating IGSCC, and Minimizing Shut Down Dose Rates, European Nuclear Conference, Nice, October, 1998.