

Key Emerging Issues and Recent Progress Related to Plant Chemistry/Corrosion in BWR Reactors

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1 Introduction

The Boiling Water Reactor (BWR) was originally designed to operate with high purity water of almost theoretical conductivity as the reactor coolant. However, during plant operation, the reactor water accumulates metallic impurities due to the very slow corrosion of BWR structural materials and due to the ingress of soluble and insoluble metallic species through the feedwater system. Some of the common metallic impurities present in the reactor water include iron, nickel, chromium, cobalt, copper and zinc, the latter two species coming from the brass condenser tubing. These impurities and other minor alloying elements arising from stainless steel and nickel alloy components accumulate on fuel and non-fuel surfaces as activated corrosion products that could result in increased radiation fields and increased occupational exposure.

Two key non-metallic ionic impurities commonly present in the BWR water include chloride and sulfate ions. Chlorides arise from condenser tube leaks or during start-up operation while sulfate arises from ion exchange resins. While the presence of very low levels of chlorides have not been an issue during normal BWR operation, the presence of sulfate ion can result in increased intergranular stress corrosion cracking (IGSCC) of structural materials at high oxidation potentials.

The high oxidation potentials arise from two undesirable non-ionic species, dissolved oxygen and hydrogen peroxide commonly present in the BWR water, as a result of the radiolysis of water. These two species increase the oxidation potential of structural materials which along with sulfate ions provide the driving force for IGSCC occurrence of reactor internals.

Therefore, the BWRs have undergone a variety of chemistry evolutions over the past few decades as a result of the need to control stress corrosion cracking of reactor internals, radiation fields and personnel exposure. Some of the key advanced chemistry changes include hydrogen addition, zinc addition, iron reduction using better filtration technologies, and more recently noble metal chemical addition to many of the modern day operating BWRs.

Thus, it is of great importance to update the BWR operators and regulators periodically on the key issues and resolutions associated with these established and evolving technologies, which is the main subject of this report. In addition, the report will also provide an update on the issues and recovery plans from the accident at Fukushima Daiichi BWRs that will include photos taken recently by the robotic cameras inside the Unit 1 BWR.

The key topics covered in the report will include the following:

- Advances in BWR water chemistry
- BWR iron reduction experiences
- BWR zinc addition experiences, fuel deposit optimization, radiation field control and occupational exposure
- Ion exchange resin issues and improved resins for efficient Co-60 removal
- BWR start-up and shutdown experiences including recent improvements
- OLNLC experiences, improvements and issues relating to ECP monitoring
- Proactive approaches to managing BWR materials degradation
- Plant specific issues associated with impurity ingress
- Latest issues and recovery plans from the Fukushima Daiichi accident

The report is expected to improve the understanding of current issues and resolutions so that informed decisions can be made by plant operators and regulators with the ultimate goal of safe and economic operation of nuclear power plants. An additional goal of the report is to provide information on the Fukushima accident situation and recovery plans so that plant operators can be more prepared proactively for the unexpected or for beyond the design basis accident situations.

2 Advances in BWR water chemistry

During the early years of Boiling Water Reactor (BWR) plant commissioning, the reactor operators faced numerous challenges to maintain plants in good operating conditions over their design life. The early operational challenges faced by the BWR operators were, increased reactor water conductivity, increased drywell or shutdown dose rates and some incidents of recirculation piping and other reactor internals cracking.

The increased reactor water conductivity has been largely due to inadequate reactor water clean-up systems, increased sulfate ion concentration due to the release of resin fines into the water, and in some cases increased chloride ion concentration in the reactor water due to condenser tube leaks. In fact, some of the component cracks may have initiated in the early years due to high reactor water conductivity as a result of the presence of these undesirable ionic species as shown in Figure 2-1 [Hettiarachchi & Weber, 2010]. However, in the later years the reactor operators have taken adequate proactive measures to lower the reactor water conductivity to almost theoretical values (after correcting for dissolved zinc ion concentration) by improving reactor water clean-up capacity and by utilizing better clean-up filters that eliminated reactor water conductivity as a factor responsible for increased component cracking propensity.

As an example towards these developments, the reactor water chloride trends at U.S. BWRs are shown in Figure 2-2. Average reactor water chloride decreased 30% from 0.40 ppb in 2000 to 0.28 ppb in 2012. Reactor water chloride at many U.S. BWRs is often reported at the lower limit of detection (LLD), around 0.3 ppb.

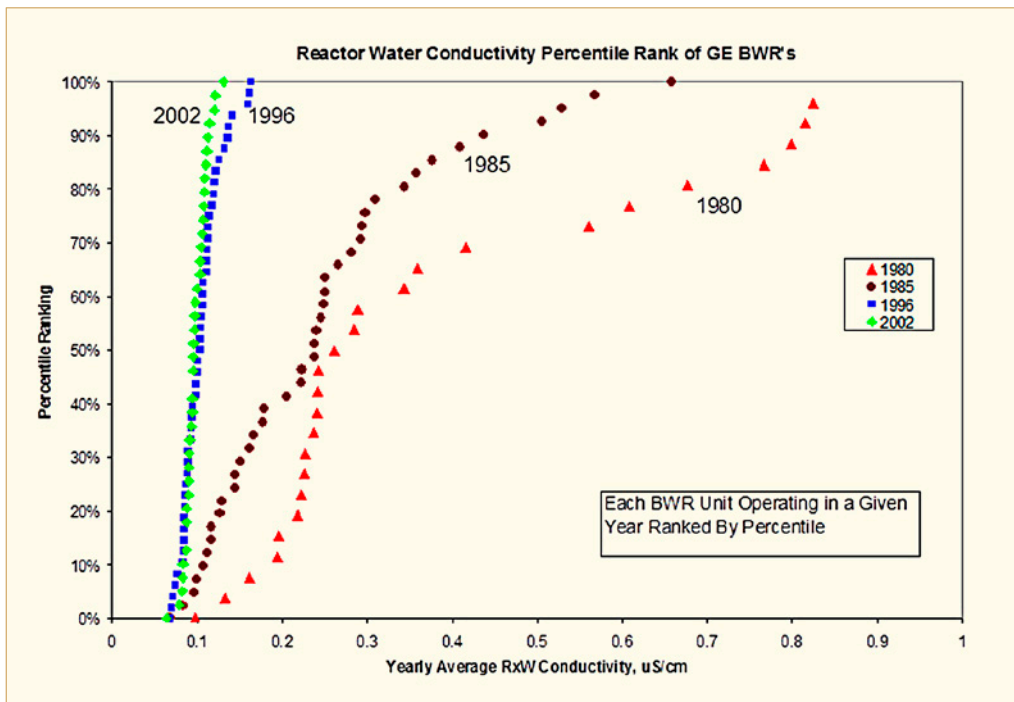


Figure 2-1: BWR reactor water conductivity improvements over the years [Hettiarachchi & Weber, 2010].

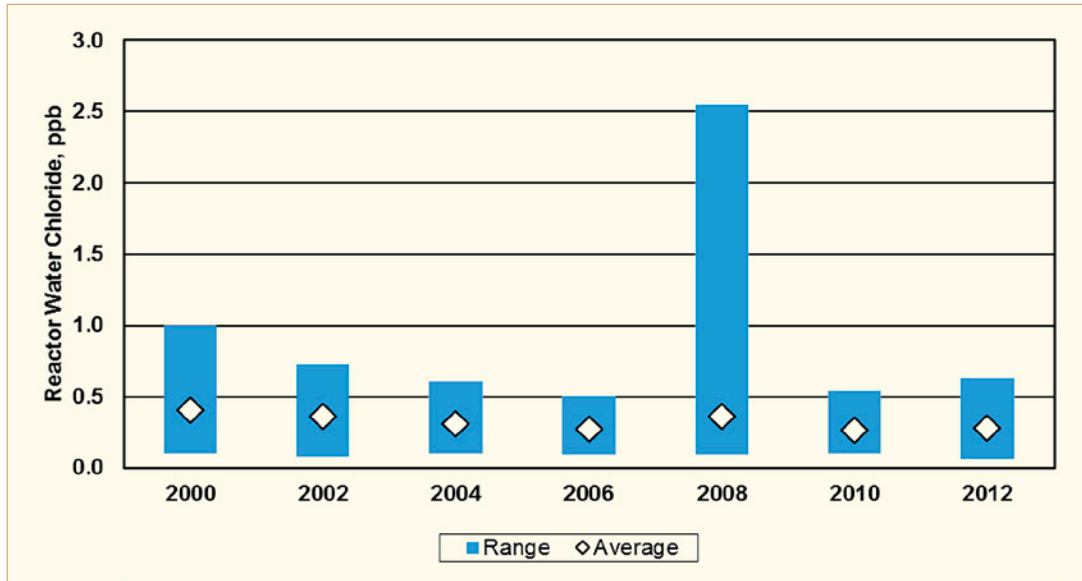


Figure 2-2: U.S. BWR Reactor Water Chloride Trends [Garcia et al, 2014a].

Average U.S. BWR reactor water sulfate decreased from 2.0 ppb in 2000 to 0.91 ppb in 2012, a 55% reduction. The improvement, shown in Figure 2-3, is particularly evident at BWRs with only deep bed condensate polishers. This is attributed primarily to the use of anion underlays and high cross-linked cation resins.

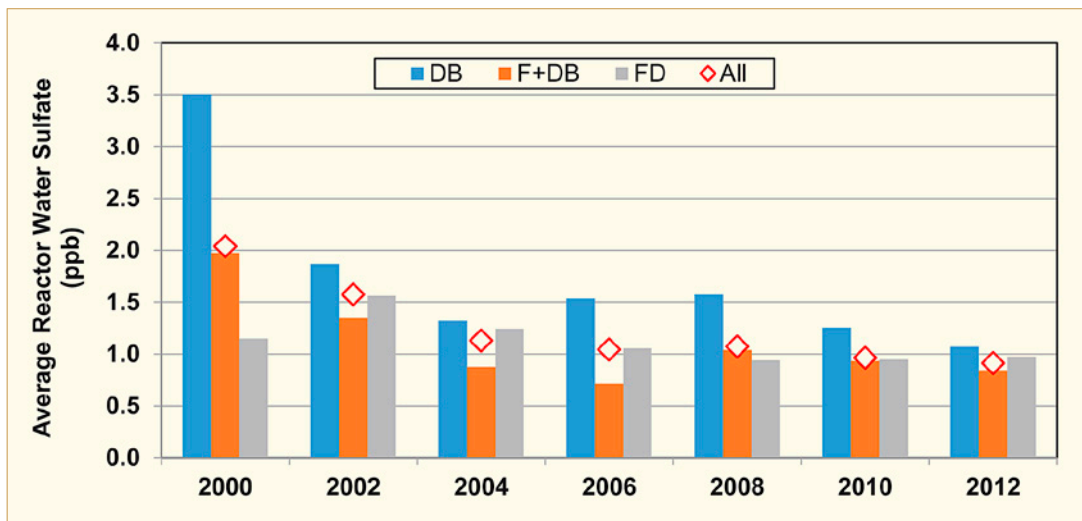


Figure 2-3: U.S. BWR Reactor Water Sulfate Trends [Garcia et al, 2014a].

It is important to optimize BWR water chemistry to reduce degradation of structural materials, maintain low radiation fields and improve fuel performance by minimizing crud formation and crud build-up.

2.1 The advent of BWR water chemistry related technologies

Along with improved reactor conductivity began the major challenge of addressing the mitigation of intergranular stress corrosion cracking (IGSCC) of structural materials. Thus, the implementation of hydrogen water chemistry (HWC) to lower the electrochemical corrosion potential (ECP) began in 1983 [Indig et al, 1985], followed by zinc addition to reduce radiation fields in 1986 [Marble, 1986]. Noble Metal Chemical Application (NMCA, i.e. addition of Pt and Rh chemical solutions) was implemented in 1996 [Hettiarachchi et al, 1995] to make HWC more effective that provided the same ECP benefit as moderate HWC (1.6 to 2 ppm H₂), but at a much lower hydrogen concentration of <0.4 ppm in the feedwater. The year 2005 saw the emergence of on-line NMCA (OLNC), i.e. the addition of a solution of Na₂Pt(OH)₆ into the feedwater of a BWR while the plant was on power operation [Hettiarachchi & Diaz, 2006], thus saving the valuable critical path time that was required with NMCA applications.

Figure 2-4 shows the application evolution of several different chemistry technologies for 48 different BWRs over the past several decades [Fruzzetti et al, 2014]. With the first OLNC application in 2005 at the Kernkraft Mühleberg BWR in Switzerland followed by the first US BWR OLNC application in 2006, the number of plants operating with OLNC+HWC has increased dramatically.

All U.S. BWRs were injecting hydrogen as of 2004. Hydrogen availability has increased significantly in the U.S., recently averaging about 98%. There are several contributing factors to the improved HWC availability trends [Garcia et al, 2014a]:

- Increased communication between Chemistry and Operations to ensure that HWC availability is maintained as high as possible
- Transition of plants to NMCA or OLNC, avoiding the need for hydrogen flow reductions for dose considerations
- Plant modifications to improve system reliability and to minimize the duration of system trips
- Plant modifications to allow HWC to be started earlier in the start-up sequence

Early hydrogen water chemistry (i.e., hydrogen addition when the reactor water reaches 200°F (93.3°C) continues to be an important area of development [Fruzzetti et al, 2014].

4 BWR zinc addition experiences

Depleted zinc oxide (DZO) addition is widely practiced across the US fleet as well as in a number of non-US BWRs. The enhanced focus on radiation reduction continues to emphasize the importance of controlling radiation source term. Shutdown chemistry control is also necessary to avoid excessive release of activated corrosion products from fuel surfaces to the coolant and to out-of-core surfaces that can delay clean-up and affect refuelling operations.

The increasing complexity of chemistry alternatives, coupled with the goals to increase output and reduce costs, continues to necessitate strategic planning to evaluate and optimize chemistry controls such as zinc addition at power plants.

Zinc continues to be used at all BWRs in the U.S., and has been effective in reducing primary system radiation fields, as measured at the BWR Radiation Level Assessment and Control (BRAC) reactor recirculation system (RRS) locations, covering BWRs 2 through BWR 6 designs for all operating chemistry regimes (NWC, HWC-M, NMCA + HWC, and OLNC + HWC).

4.1 Radiation field control in BWRs

All Exelon BWRs inject depleted zinc oxide (DZO) for radiation field control. Zinc injection rates are adjusted based on soluble Co-60 concentrations to maintain the reactor water Co-60(s) to Zn(s) ratio less than $2E-5 \mu\text{Ci/ml/ppb}$. In some cases, feedwater zinc concentrations of $>0.4 \text{ ppb}$ (cycle average) or 0.5 ppb (quarterly average), are required to meet the ratio when soluble Co-60 is elevated. Assessment of the risk to fuel reliability is considered when increasing feedwater zinc above 0.4 ppb (cycle average). The reactor water to feedwater zinc concentration factor is higher at plants with lower feedwater iron, allowing the ratio to be met at lower feedwater zinc concentrations.

Cycle median reactor water zinc concentrations and BRAC dose rates for Exelon plants are shown in Figure 4-1. Increased reactor water zinc concentrations were the result of increased feedwater zinc concentrations in an effort to reduce the reactor water Co-60(s) to Zn(s) ratio for radiation field control. The relationship between water chemistry, Co-60, feedwater iron and feedwater zinc are complex factors that are not always easy to understand.

A comparison of the BRAC (BWR Radiation Assessment Level and Control) dose rates at the selected Exelon BWRs with the median BRAC dose rate at all U.S. BWRs having refuelling outages in the same calendar year is shown in Figure 4-2. Since 2001, U.S. BRAC dose rates have been on a decreasing trend. All U.S. BWRs inject depleted zinc oxide (DZO) for control of shutdown radiation fields and operate with HWC or NMCA/OLNC+HWC for mitigation of IGSCC.

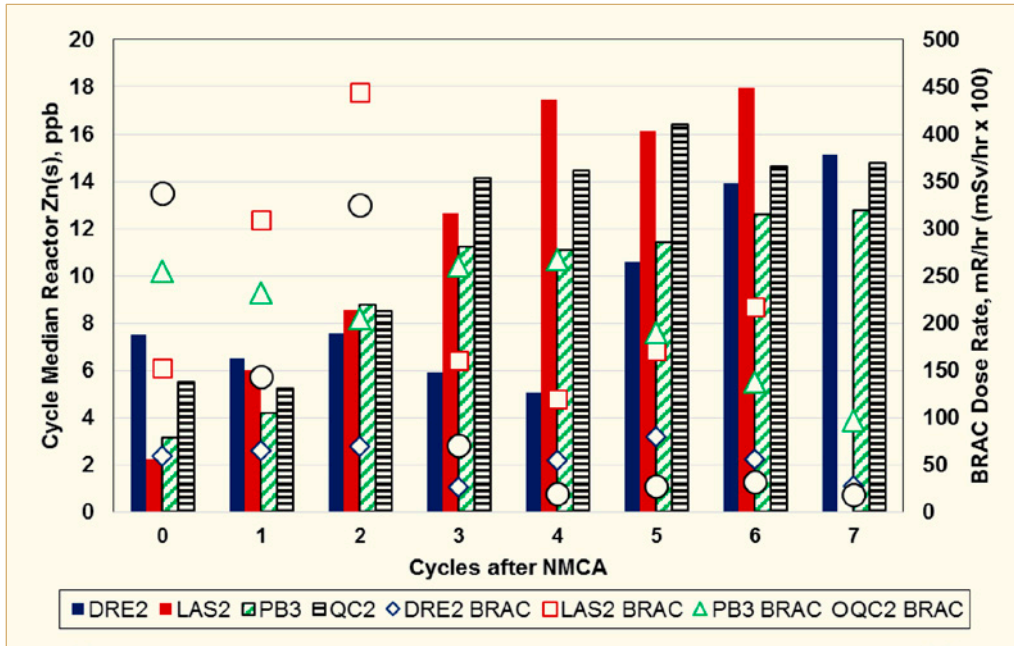


Figure 4-1: Cycle median reactor water Zinc and BRAC dose rates at selected BWRs [Odell & Jarvis, 2014].

Several studies have been conducted at Exelon Nuclear’s 14 BWRs in order to understand more completely the cause and effect relationships between reactor water radioactive species and radiation levels [Odell & Jarvis, 2014]. Most BWRs have experienced lower dose rates after On-Line NMCA (OLNC). All Exelon BWRs have made significant progress in removal of cobalt containing materials, such as original equipment manufacturer (OEM) control rod blades (CRBs) containing Stellite™ pins and rollers. The reduction in OEM CRBs at the selected BWRs is shown in Figure 4-3. All original OEM CRBs have been removed at LaSalle 2 and Quad Cities 2 as shown in the Figure.

The resulting dose rate experience of these plants following NMCA is shown Figure 4-4. The dose rates decreased with CRB replacement, increased somewhat after the 2nd NMCA application. But, all dose rates decreased following OLNC except LaSalle 2. The higher dose rate at LaSalle 2 (BWR/5) is attributed to the smaller diameter (51 cm) RRS piping having a different geometry compared to other BWRs (BWR/2 through BWR/4) that have larger diameter piping (71 cm).

6 BWR start-up and shut-down chemistry experiences

6.1 Key shutdown activities

The key process sequences to achieve cold shutdown and flood the reactor cavity include [Garcia et al, 2014e]:

- Initial power reduction to shutdown (all rods in)
- Performance of a low power scram
- Initial Cooldown
- Placing Shutdown Cooling (SDC) in service
- Reactor cavity floodup
- Post reactor cavity floodup to cavity drain

From a shutdown chemistry perspective, the overall goal is to minimize crud bursts; to minimize the amount of time required to remove activity from the reactor coolant during the shutdown process prior to refuelling, and to assist plants in predicting and controlling radiation exposure during outages. From a start-up chemistry perspective, the overall goal is to show that plants are more susceptible to IGSCC during early heat-up and start-up when reactor coolant oxidants are high in concentration. Also, during the start-up phase there is a greater likelihood for chemical excursions associated with refuel outage work activities to occur, when hydrogen injection is not available because of system design limitations [Garcia et al, 2014e].

Data from over 70 refuelling outages were evaluated to determine the Co-60 change at each of the key shutdown sequences. The data are shown in Figure 6-1 [Garcia et al, 2014e]. The plot includes all the plants that are on HWC, NMCA and OLNC. The highest change in Co-60 concentration occurs during the initial cooldown when the reactor coolant temperature is lowered from the normal operating range of 545°F (285°C) to about 350°F (177°C), just prior to the initial start of shutdown cooling, while the highest Co-60 concentration occurs just after shutdown cooling is placed in service.

For plants operating with HWC, NMCA or OLNC, by the time reactor cavity floodup commences, reactor coolant concentrations are significantly lower than the peak concentration levels. The time period between the initial start of Shutdown Cooling and the start of cavity floodup is when the reactor vessel is being disassembled. This is the period when it is important to operate the reactor water clean-up system at maximum flow and maximum availability to minimize the Co-60 concentration to as low as possible prior to flood up [Garcia et al, 2014e].

7 On-line NMCA (OLNC) experiences and improvements

This chapter deals with recent OLNC experiences that made new revelations about the Pt deposition effectiveness, radioactive corrosion product release and its origin, ECP monitoring difficulties in the mitigation monitoring system (MMS), and improvements relating to OLNC applications.

7.1 Pt deposition effectiveness of OLNC treated surfaces

The change in reactor water Pt concentration with each successive OLNC application performed at the Kernkraftwerk Mühleberg (KKM) plant is shown in Figure 7-1. The reactor water Pt concentration shows a gradual decrease, for almost the same Pt input at each cycle of operation reaching almost unmeasurably low values during the 11th OLNC application. This observation is an indication that Pt deposition on already Pt treated surfaces is more efficient appearing to catalyse the deposition process resulting in lower reactor water Pt at each successive application as seen in Figure 7-1 [Hettiarachchi & Weber, 2014].

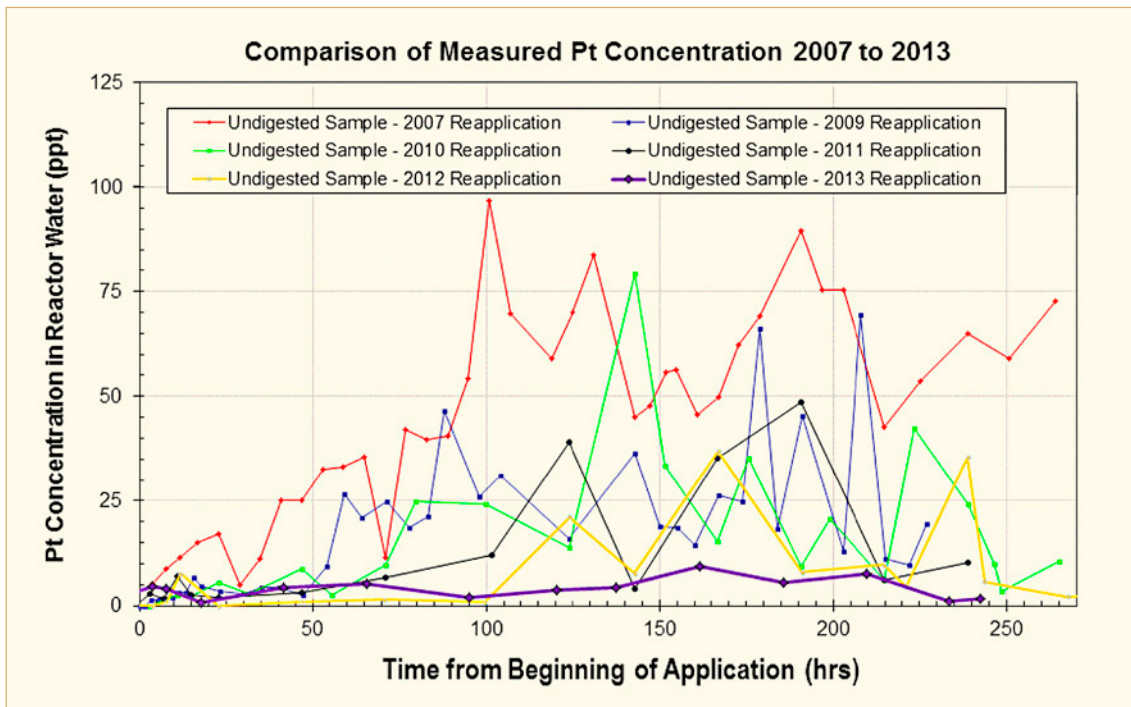


Figure 7-1: Reactor water Platinum concentration during each successive OLNC application at KKM [Hettiarachchi & Weber, 2014].

8 Plant specific issues associated with impurity ingress

BWR plants have occasionally experienced impurity ingress events due to condenser tube ruptures that have occurred under different circumstances. These events typically cause rapid increases in reactor water conductivity associated with elevated levels of chloride and sulfate ions. Since both chloride and sulfate ions at elevated temperature can enhance SCC initiation and crack growth, special attention is paid to these events and appropriate corrective action is taken immediately. The repercussions of these events are more serious if the impurity ingress is related to sea water intrusions because of the high levels of chloride and sulfate ions that can enter the reactor vessel.

8.1 Chloride intrusion event at Millstone-1 reactor

On September 1st 1972, the Millstone-1 reactor was undergoing a routine start-up. The reactor achieved less than 0.1% of full power when the operator noted a problem with the demineralizer. He switched to a second demineralizer and proceeded with the start-up. Half an hour later he noticed that there was a failure in the second demineralizer as well, so the operator began a deliberate shutdown procedure. An hour later, when excessive chlorides were noted in the primary water the deliberate shutdown procedure was abandoned and the reactor was scrammed. Sea water has intruded into the primary system of the reactor.

The ensuing water chemistry transient due to the sea water intrusion event is shown Figure 8-1. The chloride ion concentration in the reactor water reached levels close to 20,000 ppb and the reactor water conductivity reached values close to 90 $\mu\text{mho/cm}$, measured at 25°C.

An intensive investigation revealed that tubes in the condenser had corroded allowing sea water to enter the primary system. The main damage was to the local power range monitors that measures the power of the reactor. All 120 LPRMs have failed. Following this incident, all Aluminium-brass tubing in the condenser were replaced with copper-nickel tubes which have higher resistance to sea water corrosion. The LPRMs were also replaced and the reactor eventually resumed operation [Cheng, 1973].

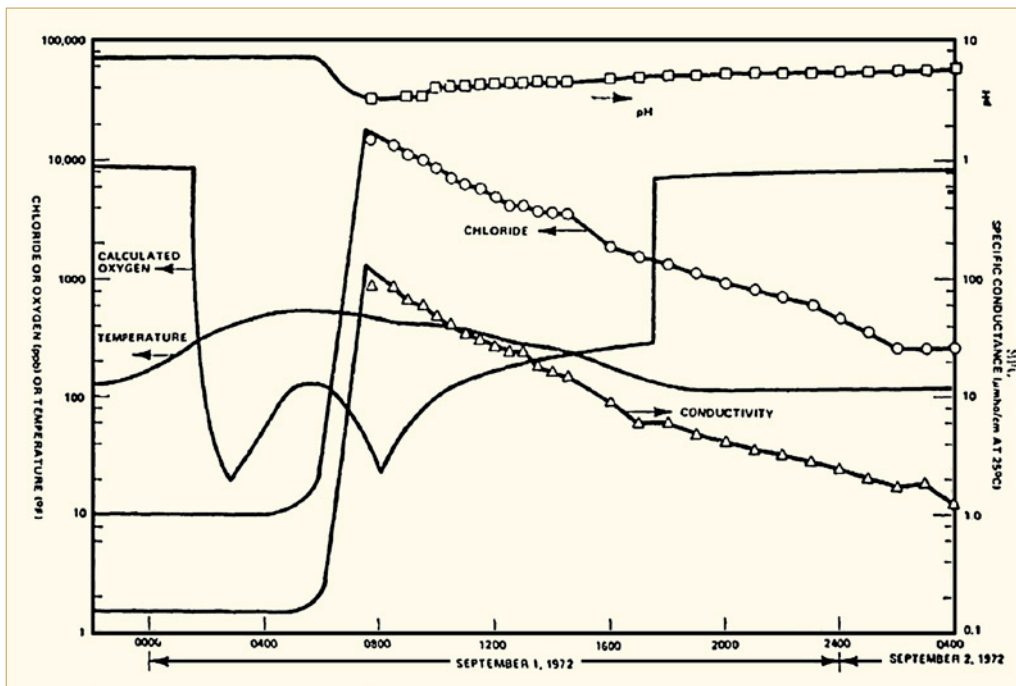


Figure 8-1: Water chemistry transient during the sea water intrusion event at Millstone-1 reactor [Cheng, 1973].

8.2 Chloride and Sulfate intrusion event at Duane Arnold reactor

In 2003, the Duane Arnold BWR experienced a 17 to 18 hour intrusion of cooling water due to a stainless steel condenser tube failure [Gordon & Garcia, 2010]. The reactor water conductivity, sulphate and chloride peaked at 232 $\mu\text{S}/\text{cm}$, 93.8 ppm and 21.4 ppm respectively during the transient event. This was the worst identified water chemistry transient in a BWR. The Duane Arnold BWR was operating under HWC and had performed NMCA application as well. What is even more important is that Duane Arnold had the only operating crack growth monitoring system in the entire BWR fleet. The monitoring system received water from the recirculating piping and was subjected to the water chemistry transient as well. The system had a furnace sensitized Type 304 stainless steel, Alloy 182 and an Alloy 600 Compact Tension (CT) crack growth specimens. The presence of these specimens allowed the real time monitoring of the impact of this severe water chemistry event on the crack growth rates of all three BWR materials of importance.

Figure 8-2 illustrates the evolution of this event. The Alloy 182 specimen had a stress intensity factor of $30.8 \text{ MPa}\sqrt{\text{m}}$. During the transient, the crack growth rate of the Alloy 182 CT specimen increased from 2.5×10^{-9} to 7.2×10^{-7} mm/s, approximately a 300 times increase in crack growth rate due to the intrusion of high levels of chlorides and sulphates into the reactor water. As the system was getting cleaned-up, the crack growth rate decreased to 1.1×10^{-8} mm/s. Calculations show that the total crack extension during the transient period was $<76 \mu\text{m}$ [Gordon & Garcia, 2010]. After clean-up, the plant returned to full power and the crack growth rate of the Alloy 182 specimen returned to its pre-transient value.

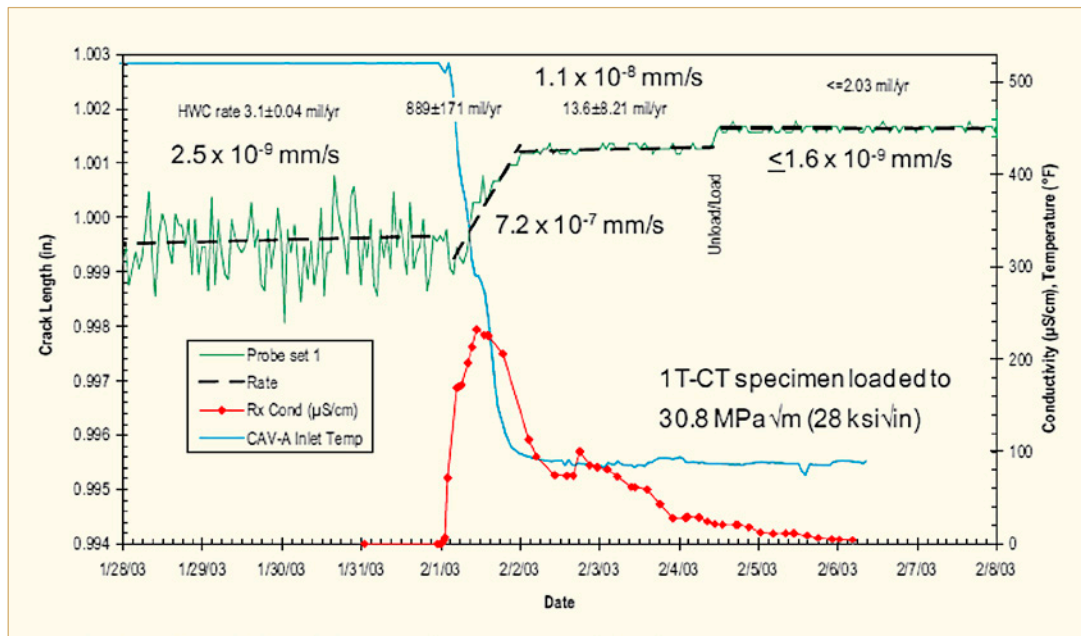


Figure 8-2: Alloy 182 crack growth rate response to the water chemistry transient at Duane Arnold BWR [Gordon & Garcia, 2010].

10 Fukushima Daiichi recovery plans and latest issues

10.1 The event and the early actions

On March 11, 2011, northeastern Japan experienced a series of huge earthquakes resulting in a tsunami that caused heavy damage to the Fukushima area and surroundings. All Units except Unit 6 lost all electricity not only from the external sources but also from the emergency diesel generators (EDGs) which were damaged by flooding of the tsunami following the earthquakes. A severe accident at the Fukushima Daiichi Nuclear Power Plant (NPP) occurred from a station blackout (SBO) due to the unexpectedly large tsunami challenging the decay heat removal from the core of the Units 1 to 3 and severe core damage, core meltdowns, leakage of the primary containment vessels (PCVs), destruction of the reactor buildings due to hydrogen explosions, and the release of radioactive fission products (FPs) into the environment [Uchida et al, 2014a]. Units 5 and 6 were safely shutdown using one of the Unit 6 EDGs that was air cooled and hence was able to supply cooling water to both Units 5 and 6 [Muto, 2014].

It was shown in the official report of the Fukushima accident that 600 PBq of FPs were released into the environment. Major parts of the FPs consisted of radioactive noble gases, *e.g.*, Xe-133 & Kr-85, radioactive iodine, *e.g.*, I-133 & I-131, and radioactive cesium, *e.g.*, Cs-134 & Cs-137. Based on previous understanding, it was considered that Cs was released from fuel into the coolant, where some of Cs reacted with iodine to produce CsI and the rest formed CsOH. The accumulated mass of Cs in the core was about ten times larger than that of iodine. Only 10% of Cs existed as CsI and 90% was CsOH in the water [Uchida et al, 2014b].

Since the accident, efforts have been made to cool down the reactors (Units 1, 2, 3) and spent fuel pools (SFPs) of Units 1 to 4 by using fresh water or sea water. Three hundred (300) tons of water is injected to cool the cores of 3 reactors [Muto, 2014]. The water injected into the reactors leaked from the damaged reactor pressure vessel (RPV) and primary containment vessel (PCV) into the basement of the buildings. A water cooling circulation system was installed that enabled the reactor temperatures of Units 1, 2, 3 to achieve <100°C and also significantly suppress the radioactive material release. The SFP water purification system was also installed to prevent corrosion and it was possible to achieve temperatures of approximately 30°C as of June 2012 [Makihira et al, 2012].

The areas affected by flooding of the Fukushima Daiichi NPP site is shown in blue in Figure 10-1.

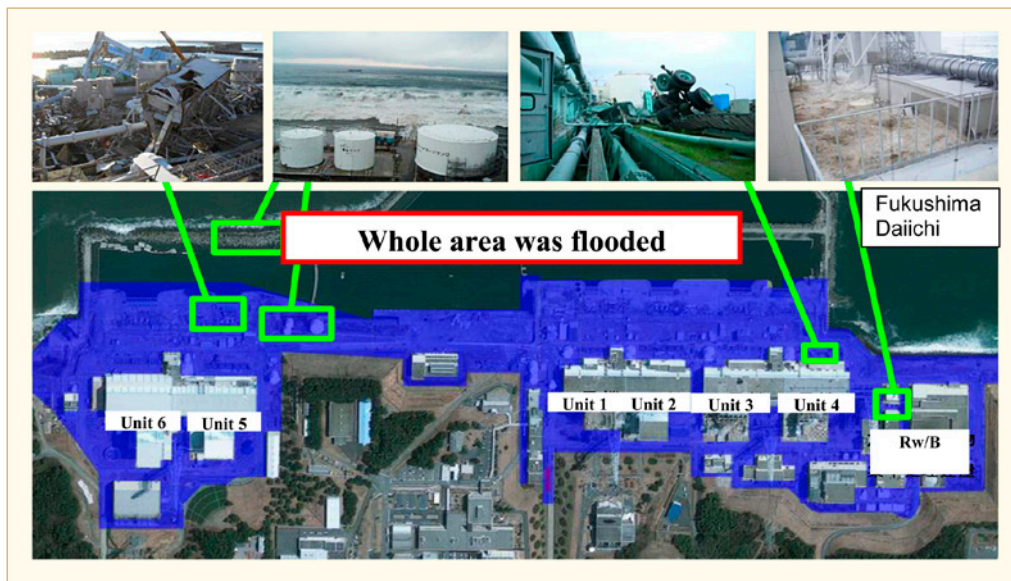


Figure 10-1: Flooded area of Fukushima Daiichi NPS shown in blue [Muto, 2014].

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