

Spring 2011

Improved Analytical Analysis of a Pressurized Water Reactor Secondary Loop

Megan J. Holba
Governors State University

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Improved Analytical Analysis
of a Pressurized Water Reactor Secondary Loop

A Project

Submitted

to

Governors State University

By

Megan J. Holba

In Partial Fulfillment of the

Requirements for the Degree

of

Masters in Science

Spring 2011

Governors State University

University Park, Illinois

Acknowledgements

I would like to thank Dr. Henne for his help and advice throughout this project as well as Dr. Addison and Professor Kent for their committee support.

I would also like to thank Laura Finney, Unit Chemist at Braidwood Nuclear Station, for providing various details on the implementation of the PAA project as well as directing and working with me throughout the analysis of the steam generator and feedwater filters. Her support and backing within our department also allowed me to pursue a better method for Fe determination.

Table of Contents

Abstract.....	3
Introduction.....	3
Pressurized Water Reactor (PWR):	3
Polyacrylic Acid (PAA):.....	5
Experimental	7
Chemicals:.....	7
Steam Generator Blowdown Filters:	7
XRF analysis:.....	7
Proposed New XRF Fe calibration:	9
Results and Discussion.....	13
BWD Percent Fe Removal:.....	13
BWD Frequency of Filter Change Out:	14
Impact on ANO-2 and McGuire 2:	15
Impact of Improved XRF Method:.....	15
Conclusion	16
References.....	16
List of Figures.....	18
Figure-1: The Pressurized Water Reactor (PWR)¹	18
Figure-2: PWR Fuel Assembly¹⁰	18
Figure-3: Cross-Section of a Steam Generator¹¹	19
Figure-4: Oxford ED2000 XRF	20
Figure-5: X-Ray Fluorescence Model¹²	20
Figure-6: Braidwood Unit 1 %Fe Removal vs. FW [PAA]	21
Figure-7: Braidwood Unit 1 FW Fe vs. FW [PAA]	21
Figure-8: McGuire 2 Iron Concentration Measurements Recorded During Long Time Trial⁵	22
Figure-9: McGuire 2 Blowdown Iron Removal Efficiency during Dispersant Trial⁵	22
Figure-10: EPRI Deposit Thermal Resistance Best-Estimate Projections for a Typical Plant⁵	23

Abstract

In the nuclear industry, the addition of poly-acrylic acid (PAA) as a dispersant to the secondary cooling loop in pressurized water reactors (PWRs) has been found beneficial in keeping the corroded Fe in solution. The purpose of this study is to measure the effectiveness of the addition of PAA to Braidwood Nuclear Power Station and other plants, to show the benefits of PAA implementation and to evaluate the current method for Fe detection. The current analytical method for the X-Ray Fluorescence Detector has an upper detection limit of 5,000 $\mu\text{g Fe}$, however, the results frequently exceed the upper limit. In order to accurately measure the amount of Fe being kept in solution, the limit of linearity needs to be researched in an attempt to maximize the dynamic range of the current instrument in place, the Oxford ED2000 XRF.

Introduction

Pressurized Water Reactor (PWR):

Pressurized water reactors (PWR) (Figure-1)¹ are a form of light water reactors that use water to cool and regulate the nuclear fission process occurring in the core. The core is comprised of fuel assemblies containing uranium dioxide pellets that are encased in zirconium alloy tubes (fuel cladding). The cladding prevents fission products from escaping the fuel assemblies, yet permits the release of neutrons, heat and other energy forms as the fission process progresses. These individual tubes of fuel and cladding are bundled together to form each one of the fuel assemblies (Figure-2), arranged to produce an even neutron flux within the reactor vessel.²

Water used as a moderator in light water reactors, removes this excess decay heat from the fission process which superheats the primary coolant (water) in the reactor vessel as this primary coolant loop is under high-pressure. The superheated water then leaves the reactor and circulates

through the steam generators, giant heat exchangers, where the high-temperature, high-pressure water can then transfer heat to a secondary cooling loop, causing the secondary loop to boil and superheat. The steam generated in this cycle is then used to turn the turbines, which rotates the main shaft, producing power in the main generator.²

This secondary loop will be the focus of this research as it is very important to minimize metal-oxide deposits from depositing in steam generators (Figure-3) to allow efficient heat transfer to the secondary cooling loop. Consequences of metal-oxide buildup in the secondary side of steam generators, resulting from the deposition of corrosion products and impurities within the water on steam generator tubes and internal surfaces, can lead to decreased heat transfer, blockage of feedwater flow and development of crevices where corrosive impurities can linger. The corrosive impurities can then lead to stress corrosion cracking then ultimately tube failure causing primary contaminated water to leak into the secondary (clean-side) water.³ In order to prevent buildup of corrosion products, PWR's primarily utilize the steam generator blowdown system in which a portion of the water from the steam generator secondary side is removed continually and sent to the blowdown system.⁴ The Braidwood Station steam generator blowdown system is comprised of pre-filters in order to remove non-soluble corrosion products and then mixed bed demineralizers to remove the soluble impurities. During operation, the blowdown system typically removes a mere 10% of both metal-oxide and soluble impurities that enter the steam generators, with the remaining impurities left to deposit in the secondary side throughout the fuel cycle. To counteract the deposition of impurities, there are many chemical and mechanical methods employed during outages.⁴

With 18 month fuel cycles utilized at Braidwood, the outages are used to make necessary repairs and cleanings of essential systems including the steam generators. Steam generators

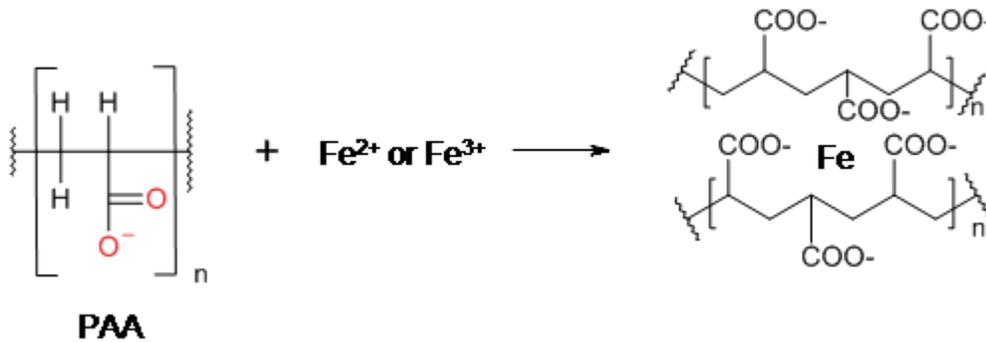
clean-up methods include sludge lancing, bundle flushing, crevice flushing, chemical soaks, pressure-pulse cleaning and chemically enhanced pressure-pulse cleaning all of which are very costly in the hundreds of thousands of dollar to millions of dollar ranges and result in increased dose to personnel involved. For example, sludge lancing is performed every fuel cycle to remove approximately two percent of the total corrosion products entering the steam generators over a typical 18 month PWR fuel cycle, costing approximately \$350,000 in 2001. True chemical cleaning of the steam generators is a very time consuming and expensive process that removes virtually all the corrosion products in the steam generators, which is required at least once in the lifetime of the steam generators and costs \$5-\$10 million per cleaning (2001 costs). These processes are attempts to remove the excess oxide and corrosive deposits that build up throughout the fuel cycle; however they are extremely costly and add to the downtime of the reactor.⁴ Every day one of the reactors is offline and not making power results in a loss of roughly a million dollars of revenue per day, therefore it is in the best interest to employ another technique to reduce the amount of corrosion product deposition in the steam generators.

Polyacrylic Acid (PAA):

Another technique developed in the last few years to reduce the amount of corrosion product deposition is the addition of a polymer dispersant. This polymer dispersant program was developed by ComEd in conjunction with BetzDearborn in the 1990's to utilize a high molecular weight, ultra-pure polyacrylic acid (PAA) as a dispersant for corrosion products in the secondary side. In the late 90's the program was turned over to the Electric Power Research Institute (EPRI) where a three month trial was performed at Arkansas Nuclear One, Unit 2 (ANO-2) in order to demonstrate PAA's ability to increase the efficiency of blowdown Fe removal in a field setting as opposed to laboratory setting. Following the successful short-term trial and calculated

benefits of PAA addition at ANO-2, a 9 month longer trial was performed by Duke Power at their McGuire plant on unit 2 (McGuire 2).⁵

Optisperse PWR6600 (approximately 10% aqueous PAA solution) provided by GE Betz, Inc., is a long chain, high molecular-weight polymer dispersant (~70,000 – 150,000 AMU⁴), that promotes the suspension of insoluble corrosion products by placing a surface charge on the iron oxide particulates promoting repulsion from one another.⁶ The carboxylic acid groups on the PAA bind to the surface of the iron oxide particles causing electrostatic and steric repulsion preventing corrosion product particle aggregation.⁷



The iron that is suspended by the dispersant can now remain in solution until it is removed via the steam generator blowdown system. Not only does PAA increase the iron removal of current corrosion products, but it can also liberate previous depositions of iron in the steam generators and the secondary cooling side. This deposition of the iron in the steam generators is greatly reduced by addition of very low concentrations of the PAA dispersant (typically 0.1 – 5ppb) throughout each fuel cycle in a PWR. As iron is suspended in the secondary side cooling, the steam generator blowdown filters should foul more quickly and need more frequent replacing, especially during the initial startup of the PAA system as the iron removal rate is expected to increase substantially.⁶

Experimental

Chemicals:

Optisperse PWR6600 provided by GE Betz, Inc. is comprised primarily of 10% aqueous PAA and monoethanolamine (ETA) as an acid neutralizer (8.7% nominal range) to boost the pH to a nominal value of 8-9 S.U.⁶ Other impurities listed include K, P, Cl, Na, F, SO_4^{2-} , and Silicon which have detrimental effects on the chemistry of the secondary side, therefore GE Betz has purified the chemical to limit the impurities to <10ppb so that once injected into the system the concentrations seen in blowdown are <1ppb in order to mitigate any adverse effects from these impurities.⁴

Steam Generator Blowdown Filters:

The Braidwood Nuclear Power Station and typical steam generator blowdown (SGBD) systems are comprised of pre-filters in order to remove non-soluble corrosion products and then mixed bed demineralizers to remove the soluble impurities.⁴ Braidwood used to employ SGBD prefilters with a 10 μm absolute pore size to eliminate the non-soluble corrosion products; however, with the addition of PAA, the station has switched to prefilters with a 5 μm absolute pore size, which has shown to more effectively remove the dispersed Fe. The mixed bed demineralizers are loaded with 60ft³ of mixed bed resin and then topped with an additional 60ft³ of cation resin for increased efficiency for the removal of amine products and Na.

XRF analysis:

In order to measure the effectiveness of Fe suspension due to the addition of PAA and the percent Fe removal efficiency, samples are taken of the water leaving the steam generators, particularly the 1A SG, and another sample is taken downstream of the SGBD filters, recorded as

unit 1 feedwater (1FW). Sample streams are run in to integrated samplers through two paper filters; one 0.45 μ m pore size particulate filter and a cation filter. These filters are then analyzed on the Oxford ED2000 XRF (Figure-4).

The Oxford ED2000 includes a floor-standing XRF spectrometer and PC. To quantify many elements, the instrument utilizes the principles of energy dispersive X-ray fluorescence (EDXRF) and Oxford XpertEase software. Liquid nitrogen cools the detector to decrease electronic noise with an internal 10L dewar. The sample chamber allows multiple sample types to be accommodated; however, in the nuclear industry, an eight-position sample tray is utilized to center our corrosion sample filters over the circular aperture in the back of the sample changer.⁸

X-Ray Fluorescence involves an X-ray of sufficient energy striking an atom and dislodging an inner electron from the K or L shell. In order to stabilize the atom, an electron from an outer orbital (the L or M shell) fills the vacancy in the inner shell, releasing excess energy in the form of an X-Ray photon (Figure-5). Since each atom's electrons' quantum states are unique, the energy released characterizes a particular element. The number of photons released at specific energies is also proportional to the concentration of the element in the sample being analyzed. One benefit to using XRF is that the peaks observed result from the ejection of inner shell electron, not bonding electrons; therefore, the energies released are independent of the chemical form of the element. Two of the main advantages of XRF are the speed of analysis and the minimal sample preparation. Typically, measurement times of 1-2 minutes provide a good and accurate signal-to-noise ratio.⁹

The elements of interest in this application are Fe, Cu & Zn, considered medium elements that emit K-lines when they undergo XRF. Fe emits the lowest energy line at 6.39keV with the

highest energy edge at 7.11keV. Cu emits the lowest energy line at 8.02keV with the highest energy edge at 8.98keV. Finally, Zn emits the lowest energy line at 8.61keV with the highest energy edge at 9.66keV.⁸ The current calibration employed on the XRF analyzes for Fe, Cu & Zn with an upper limit of 5,000µg. Due to the addition of PAA, the Fe removal efficiency has greatly increased leading to many of the sample filters being over-range on Fe for the detector. Normally, filters are installed in integrated corrosion product samplers with a continuous flow of the sample stream over a period of 3-4 days, resulting in total throughput of approximately 400-700L. After the implementation of the PAA program, the sample for 1A SG filter throughput has now been reduced to only 2L in order to keep within the limitations of the current Fe calibration.

Proposed New XRF Fe calibration:

After comparison with other nuclear sites, most sites had upper calibration limits similar to Braidwood Station; however, one station had claimed to have a linear calibration that would measure up to 40,000µg Fe, evidence that there is room to increase the current upper limit for the Fe calibration at Braidwood. New calibration standards will be ordered ranging up to 40,000µg of Fe to test the limit of linearity. Modifications can be made at a later date and or the upper calibration standards can be removed if the calibration curve proves not to be linear. There are three steps involved for full method development:

1. Define Method Parameters
2. Measure the Standards
3. Carrying Out and Saving a Regression

In order to perform a quantitative analysis calibration, select **Method Setup** from the main menu, click the **Method** button, then click **Create a New Method** in the calibration section of

the Method Editing screen. First, the name for the new method must be entered and press the **Continue** button. The next screen displays a list of elements available to scan. After selecting Fe, Cu and Zn, select the **Next** button. Next, the units and decimal places for the new method need to be set & applied. **Next** should then be selected to proceed to the “Select other Elements for [Method]”, which allows the user to select any possible interfering elements with overlapping energy lines to the analytes of interest.⁸

The “Select Fixed Conditions” screen will then be available to select either one or up to five fixed conditions (tube voltage, energy range, etc.) to optimize the measurement of the intended analytes. After matching the current Fe, Ni & Zn method parameters to the new calibration, **Next** should be selected. A specific X-ray tube current and live-time must be set for each fixed condition before selecting **Next**. A profile or region of interest (ROI) must be specified for each “analyte” (A) or “other” (O) element in the “Analyte Profiles/ROI Method” screen. Analytes typically have profiles assigned, therefore click on the intended analyte to select and then select **Profile**. Make sure the “Match Profiles to FCD” option is selected so that only those profiles that match the fixed conditions previously set will be shown. This process should continue until each analyte or other element has either a profile or a ROI assigned to it. More than one profile or ROI may be assigned by using the “Split Analyte” option.⁸

Next the “Summary of Method” screen will display all the analytes, the profile/ROI assigned, the type of calibration and the current status of the method. “CPS Only” reveals that only the analytical parameters have been defined and the method is not complete, or “Complete” status indicates that all steps are complete. The **Back** button may be utilized to change any of the method settings or **Next** can be satisfied with the current method.⁸

The “Method Options” screen allows the user to set the printer, save options and select the box for the sample spinner. Click **Save** and then **Yes** to answer if setting up samples (SUS) are to be specified for the calibration of the new method. The SUS’s which will be used for restandardization and calibration intervals are set in the “Specify SUS” screen. To do this, highlight the desired analyte and select the **Add New SUS** button so the “Select Standard” window is displayed. Select the proper SUS and in the “Available Lines” box will be a list of elements contained in the SUS that can be measured under the current method and in restandardization. Select the energy line needed to correct the element in the method and the “High/Low SUS” option in order to remove the background of the spectra leaving just the net intensity. Each SUS can then be assigned to an analyte by clicking **Apply**.⁸

Restandardization can be set at intervals or by simply monitoring the quality control (QC) check. After all SUS and restandardization intervals have been set, click **Ok** then **Yes** to save changes to proceed to the “Method Editor” screen. A brief summary of the method will be shown and **Exit** can be selected to return to the “Method Setup” screen, where a series of standards can now be measured via the “Measure Standards” option to perform a regression on a calibration line.⁸

After the **Measure Standards** button is selected, click **Select Method** to show all current methods. The new Fe, Cu & Zn method should be selected and then click **Ok** to display the new calibration box. For first time calibrations, select **Ok**, then **Yes** to enter data for the standards in the “Standard Name” window. Enter the appropriate calibration standard information, and then click **Next** to proceed to the “Concentration” window, where the concentration of the standard will be entered, then click **Next**. These steps will be repeated for all calibration standards, and then the information will be saved after the input of the final standard. Select **No** to enter

another standard and the “Master Standards Editor” screen will appear showing all the standards listed with their corresponding concentrations.⁸

Select **Exit** to move on to the “Add Calibration Standard” screen so that each of the standards can be specified to be used for the calibration and their corresponding tray position identified. Prior to selecting the standards, ensure the “Match Standard Elements to Method” box is checked to ensure the standards containing the elements of interest are displayed. Click the first calibration standard, then the page arrows to designate which tray position the standard is located, clicking **Add** to enter the standard into the tray. Continue until all standards are assigned positions and select **Ok**. The “Multi-Sample Analysis” screen will then show the standards and method name. Click **Start Run** to analyze standards. Select **Exit** after all standards have been measured and then **Yes** in order to perform a regression.⁸

If done immediately after running the calibration standards, the “Select Method” screen will appear. Highlight the method to display the “PCXRF Regression” screen showing the results of the first analytes straight line regression. The regression details include range, standard error, degrees of freedom and sensitivity. If more than one regression has been performed for the analyte, highlight the best regression otherwise the default is the most current regression. Click the **Current** button to view the “Regression Summary” screen and then click the down arrow next to the working analyte to view the entire list of available analytes. In order to apply the regression changes to all analytes, select **All**, otherwise the regression will only apply to the analyte selected.⁸ These instructions will provide the means to investigate the possibility of increasing the upper limit the calibration.

Results and Discussion

BWD Percent Fe Removal:

Not only does PAA increase the iron removal of current corrosion products, it can also liberate previous depositions of iron in the steam generators and the secondary cooling side. This deposition of the iron in the steam generators is greatly reduced by addition of very low concentrations of the PAA dispersant (typically 0.1 – 5ppb) throughout each fuel cycle in a PWR. As iron is suspended in the secondary side cooling, the steam generator blowdown filters should foul more quickly and need more frequent replacing, especially during the initial startup of the PAA system as the iron removal rate is expected to increase substantially.⁶ For Braidwood Station Unit 1, the PAA program was implemented in January 2010 at a cost to the station of \$25,000 per year.

The U1 PAA addition started at a very low concentration 0.1ppb and throughout the cycle slowly increased to approximately 1.0ppb. As shown in Figure-6, the addition of PAA initially caused a slight increase in the percent Fe removed in January and February. The increase to 0.3-0.4ppb PAA during March 2010 and then the increase to 0.5ppb PAA in late May 2010 caused a major increase in the amount of Fe removed from the secondary system. With the percent Fe removal ranging up to 450%, the data show that not only are the current corrosion products being kept in solution but the previous depositions are being removed as well. The percent Fe removal data does show that there was an initial spike and then it seemed to level off prior to the refuel outage in October & November 2010 when all systems were shut down. As shown in Figure-7, the rate of FW Fe should remain unchanged due to the addition of PAA showing the SGBD filters are effectively removing the Fe in suspension.

EPRI PWR secondary water chemistry guidelines require all PWRs to measure FW Fe concentrations at least weekly using integrated samplers, which is pertinent to assessing the effectiveness of the PAA addition. Blowdown iron removal efficiency is best directly measured by comparing FW and blowdown iron concentrations. Determining the blowdown iron removal efficiencies is plant specific and not required, but is beneficial in the determination of the optimal long-term FW PAA concentration and is a way to quantify the amount of Fe removed from the SG's. The amount of Fe removed from the SG's will in turn provide data to show the benefit of the PAA dispersant program as well a better estimate of the SG Fe deposition leading to more accurate prediction models that plants use to budget SG cleanings in refuel cycles.⁵

BWD Frequency of Filter Change Out:

During the initial startup, the SGBD prefilters were changed out at an increased frequency and now have tapered off, due to the leveling off of SG Fe in solution. There was no observed increase in the frequency that the SGBD demineralizers were changed out as compared to the frequency prior to PAA injection. Due to the rates at which Fe is being removed from the secondary system, sludge lancing the Unit 1 steam generators has now been decreased from every fuel cycle (18 months) to every other fuel cycle, resulting in a savings of approximately \$250,000 - \$300,000. A big concern with the deposition of Fe in the steam generators is a decrease in heat transfer efficiency resulting in a decrease of Megawatts/hour generated from each unit. Braidwood's status prior to PAA addition was that the plant was trending that way, with no loss yet but it was imminent. However, the trend seems to be leveling off with hopes that the plant will trend in the positive direction in the future.

Impact on ANO-2 and McGuire 2:

ANO-2 blowdown Fe removal efficiencies were also seen greater than 100% for the short three month trial. The SG and FW Fe data was very scattered due to the short trial period, therefore no trending can be observed. Despite the difficulties in trending, the SGBD Fe removal rates did see an increase in approximately 10% compared with the measurements prior to the PAA addition.⁵

McGuire 2 observed the same initial injection of dispersant resulted in a spike of SGBD Fe removal efficiency (Figure-8) as ANO-2 and BWD 1. This observation implied that not only the Fe in solution was being removed but the loosely adhered Fe that had settled out in the steam generators and secondary side was slowly being cleaned up as well. This trial was also implemented long enough to determine that injection rates of PAA 2-3 times more than that of FW Fe concentration resulted in SGBD Fe removal efficiency of 45-50% compared to only 6% removal efficiency (Figure-9) prior to PAA addition.⁵

Impact of Improved XRF Method:

The method on how to implement a new calibration for the XRF Fe detection has been defined and the standards are ordered so that one can now test the new method. The new method is promising and the data from other plants suggest that BWD can at least increase the upper limit of detection to 10,000µg Fe with the hope that we can achieve a linear calibration with an upper limit of 40,000µg Fe. By increasing the upper limit, more SG Fe corrosion filter papers will be in range and we will be able to have more than two liters filtered through, leading to increased reducibility and decreased source of error to support our increasing need for accurate Fe analysis.

Conclusion

Consequences resulting from corrosion product buildup in the secondary cooling loop of PWR's include steam generator tube blockage, reduced steam output due to inefficient heat transfer in the steam generators, loss of electrical output and eventually total replacement of the steam generators.⁴ All of these consequences are quite costly with the largest being total steam generator replacement, a cost that totaled \$300 million in 2009 to the Three Mile Island plant. With the implementation of the PAA dispersant program, Braidwood has seen an increase in Fe removal efficiencies. EPRI comprised a best-estimate graph revealing a moderate predicted heat-transfer benefit with dispersant use (Figure-10).⁵ The increased removal rates are promising in hopes that not only will this program combat the yearly costs of steam generator cleanup via sludge lances but also delay the replacement of steam generators all together, as shown in the EPRI documentation. The new XRF method looks to be very promising as to at least offer greater reproducibility of results, ability to analyze filters with greater throughput and give more accurate numbers with fewer filters measuring in the over range category.

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List of Figures

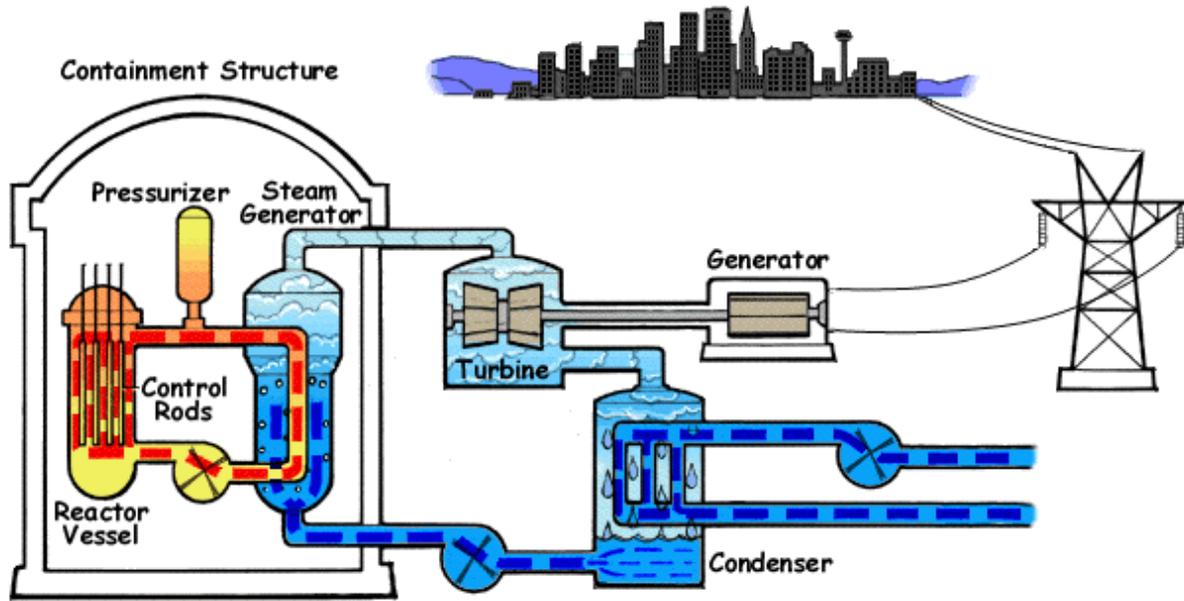


Figure-1: The Pressurized Water Reactor (PWR)¹

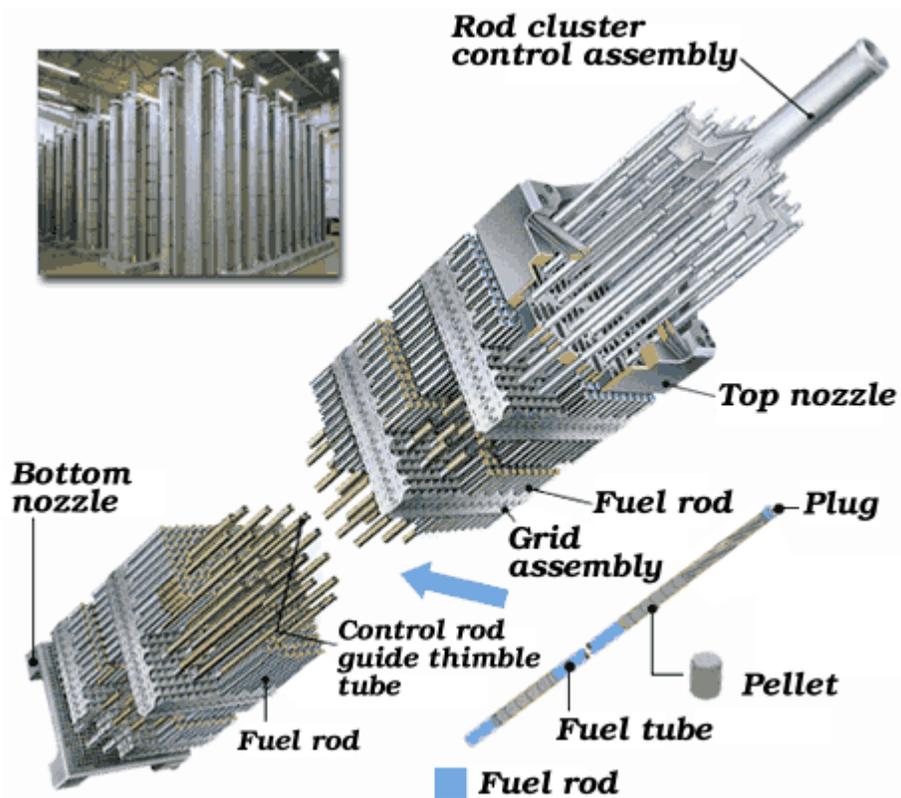


Figure-2: PWR Fuel Assembly¹⁰

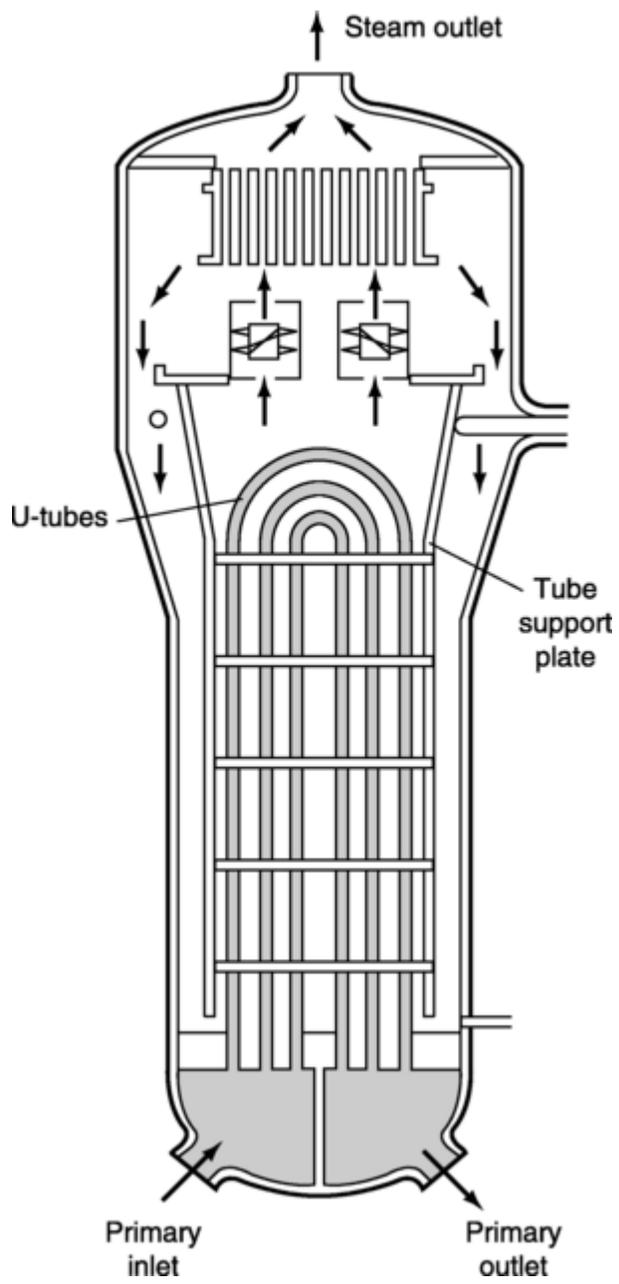


Figure-3: Cross-Section of a Steam Generator¹¹



Figure-4: Oxford ED2000 XRF

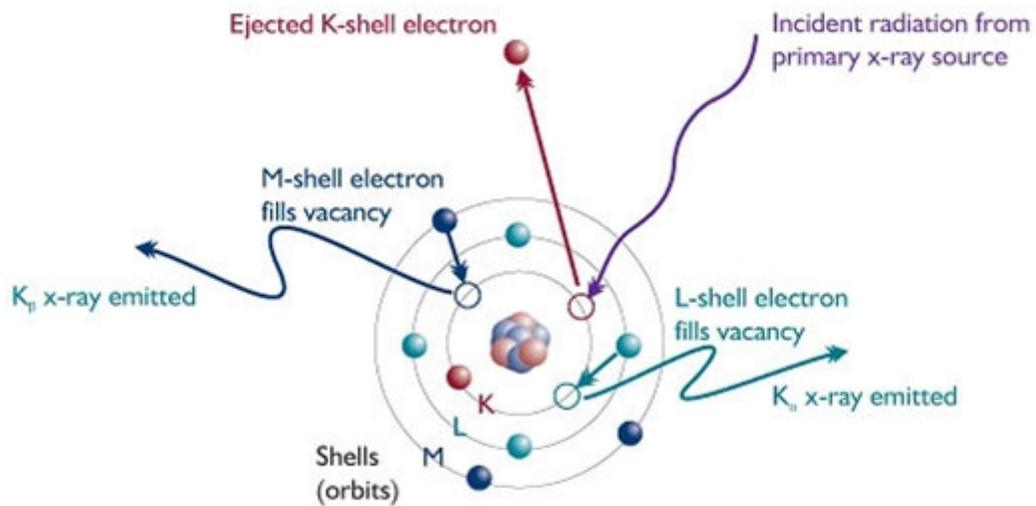


Figure-5: X-Ray Fluorescence Model¹²

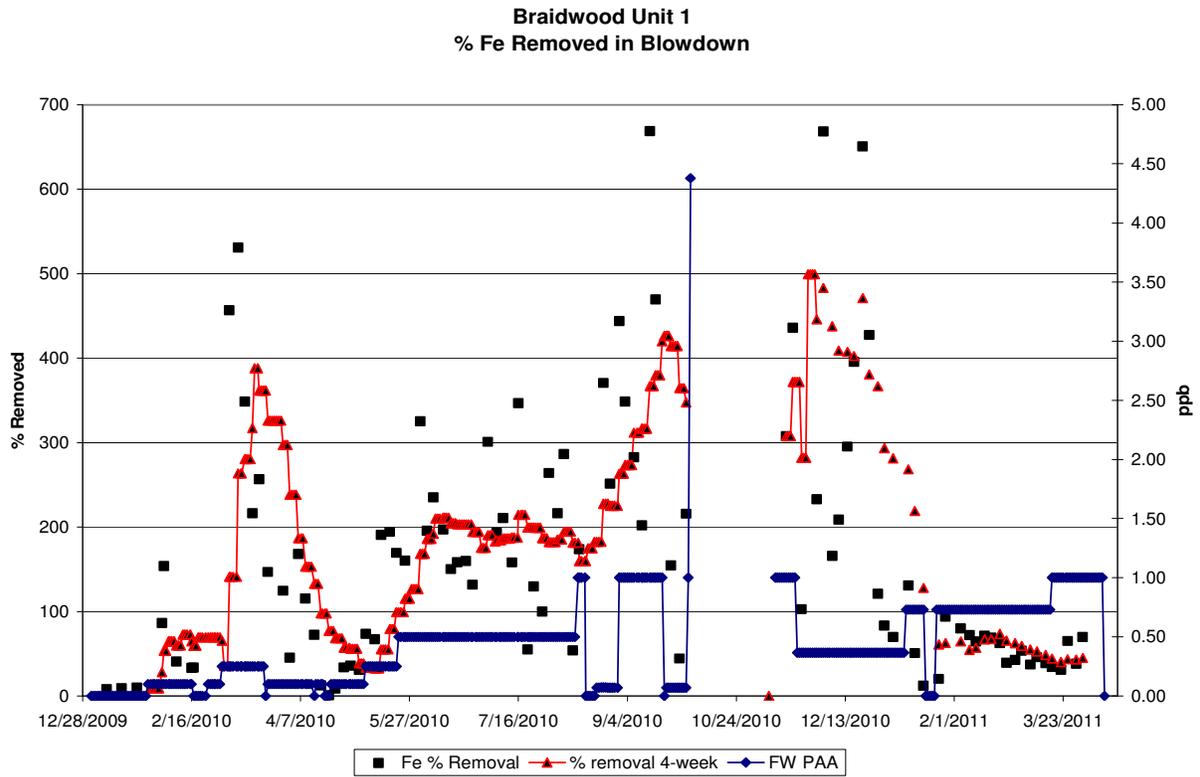


Figure-6: Braidwood Unit 1 % Fe Removal vs. FW [PAA]

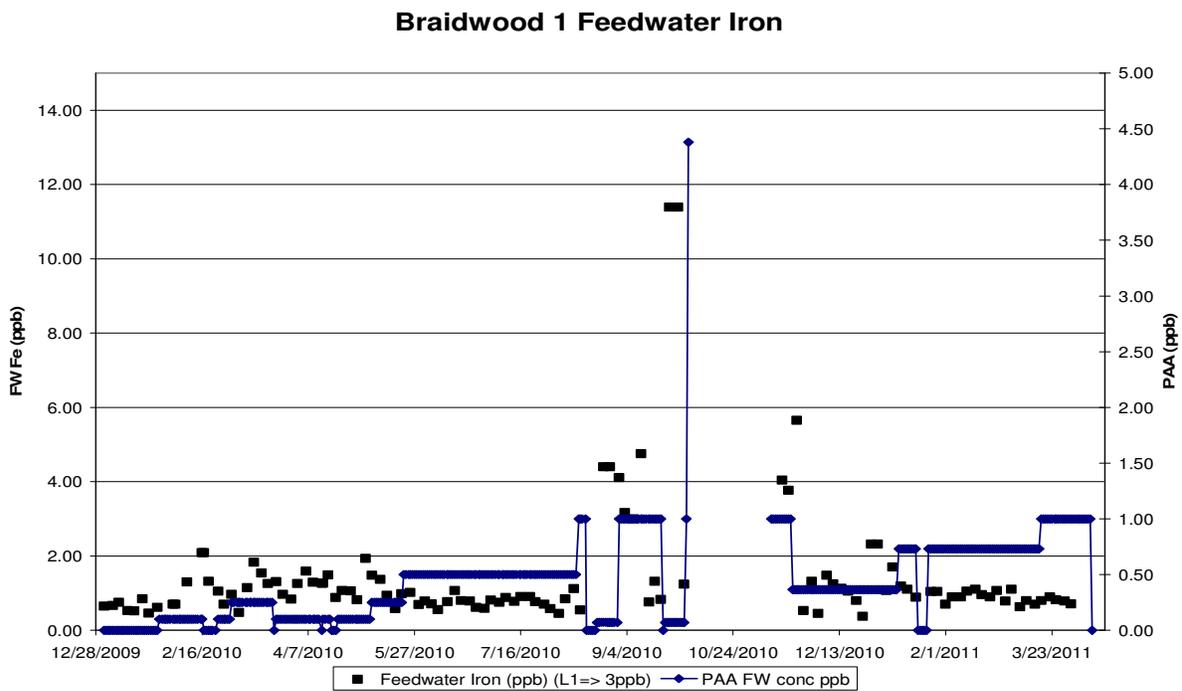


Figure-7: Braidwood Unit 1 FW Fe vs. FW [PAA]

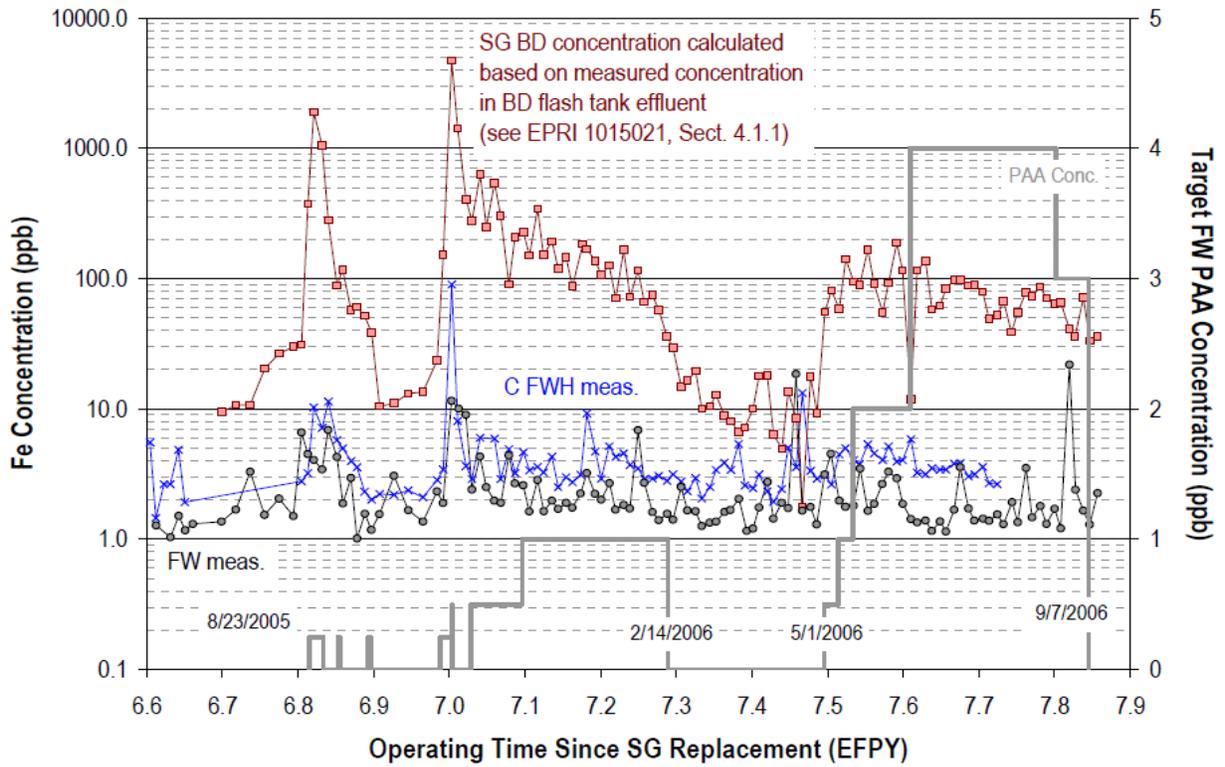


Figure-8: McGuire 2 Iron Concentration Measurements Recorded During Long Time Trial⁵

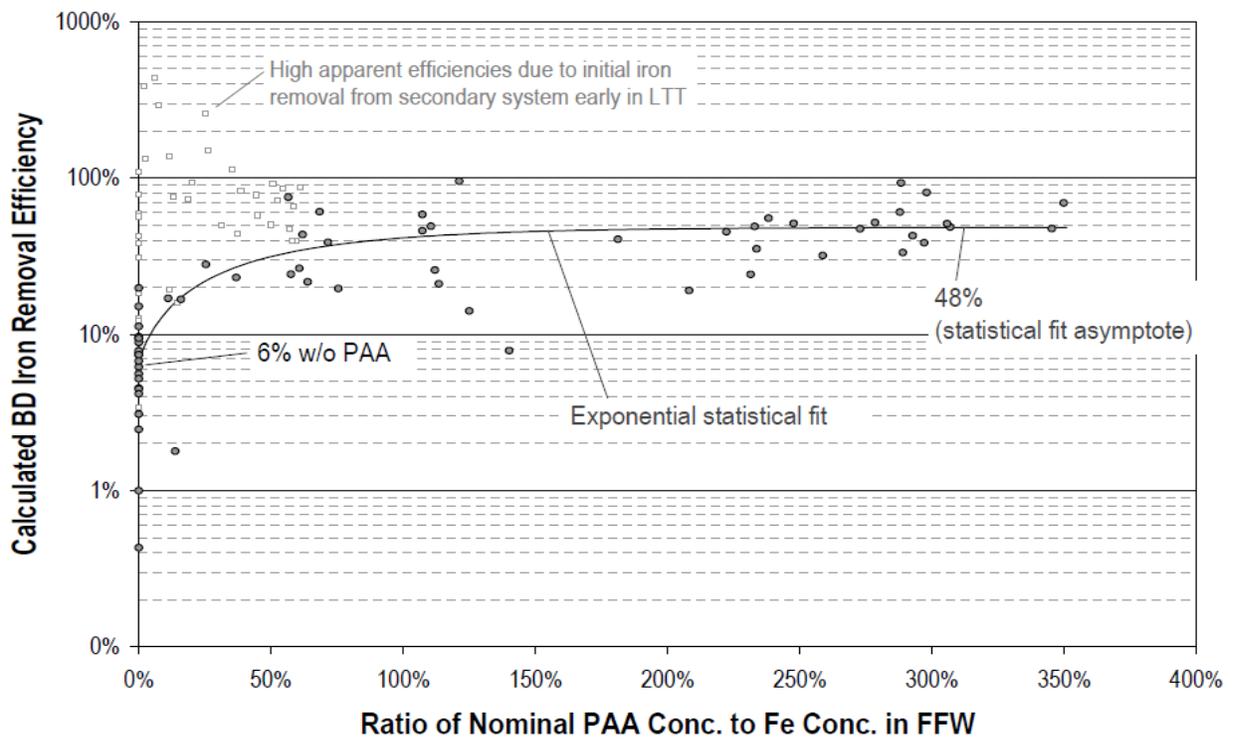


Figure-9: McGuire 2 Blowdown Iron Removal Efficiency during Dispersant Trial⁵

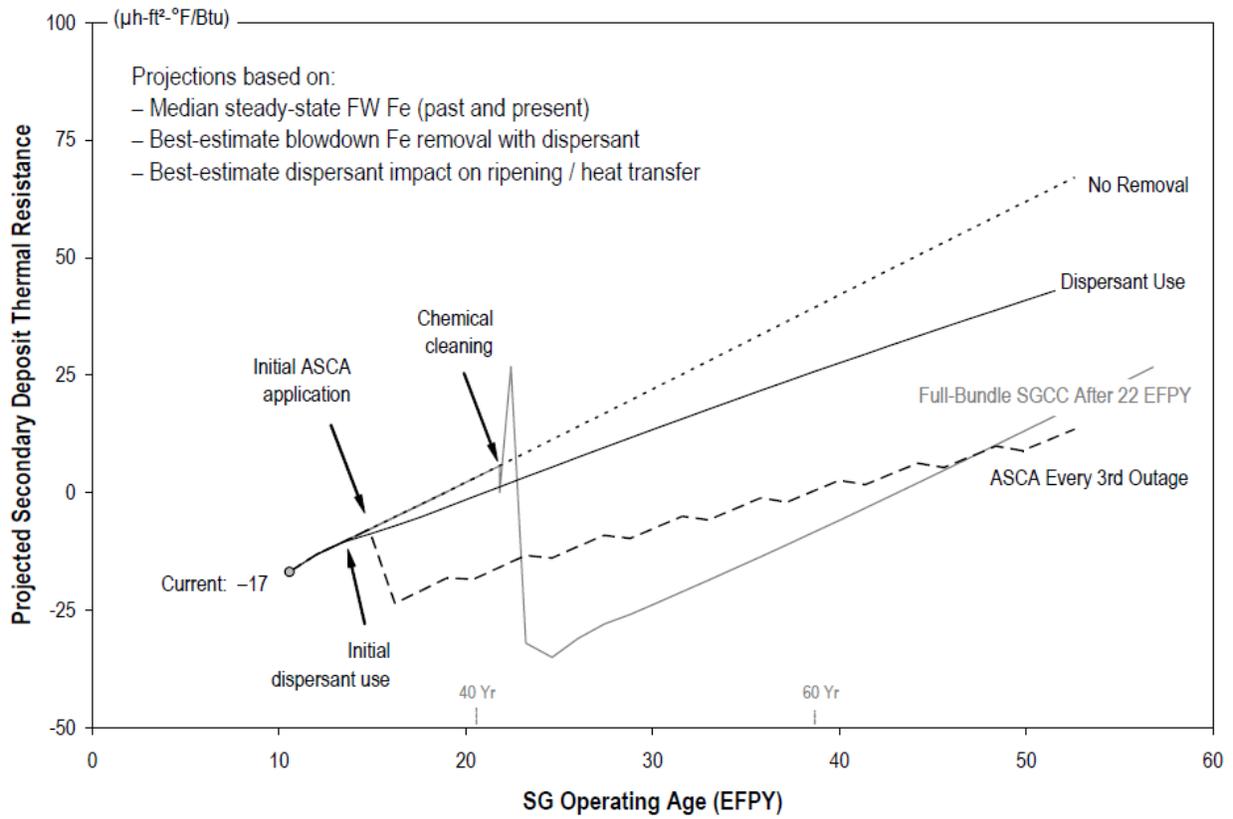


Figure-10: EPRI Deposit Thermal Resistance Best-Estimate Projections for a Typical Plant⁵