Spent Fuel Pools At Fukushima; Follow On Report "Corrosion"

by Dean Wilkie

* A companion report that explains the basics and terminology of corrosion as it relates to Fukushima Daiichi can be found here. [Corrosion At Fukushima Daiichi Explained](#) (a copy also resides further in this PDF document)

This is a follow up to a report that was issued concerning the Fukushima spent fuel pools. The first report captured general design, storage and water quality necessary to protect the spent fuel for re-use or eventual storage in dry casks. At the time the report was written the details of water chemistry within the SFP’s was not known in detail which would have been necessary to more accurately predict corrosion for the spent fuel elements, the structural racks, and the spent fuel liner/interface with the reactor wells. We have witnessed several conditions in or around the spent fuel pools since the first report. Foreign material have been introduced into the pools as a result of cleanup on the main floor area, covering spent fuel pool #4 and dropping a large steel beam into the unit 3 SFP. Cooling systems to the SFPs have been shut off many times to make mechanical adjustments causing fluctuations in pool water temperature.

Chloride levels in the SFP’s were extremely high (in the 6900’s ppm as opposed to normal of 30 ppm or less) and were left unchecked for months. (10) Every worst case corrosion analysis assumes Chloride levels at least a factor of 10 lower for their extreme conditions. Some corrosion rates have gone from 1mm to 10mm/yr which would suggest potential corrosion of the structural parts of the fuel elements (lattice holders, springs, handling assemblies, pellet holding tubing and fuel pellet cladding. The spent fuel pools have had so many foreign materials introduced that it is very difficult to pinpoint the chemical makeup of the water. Season conditions around Fukushima have contributed to the addition of dust and other foreign materials which can change the chemistry in the pools. The following parameters were mentioned in the earlier report:

- Ph
- Conductivity
- Total Oxygen
- Dissolved Oxygen
- Algae/Slime Control
- Biological Fouling
- Filterable Solids
- Temperature
- Boron
- Chlorides
- Sulfate
- Iron
- Microbial
- Biofilms

An IAEA document (1) examined the storage of fuel elements in spent fuel pools for short and long term storage. The following examples of water chemistry in storage pools is given below:

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<thead>
<tr>
<th>Parameter</th>
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<td>Cs137</td>
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“Impurities in the water of the basins used for fuel decay or interim storage can have several consequences. Aggressive ionic species, undissolved particulates (if settled on the fuel cladding), and several microorganisms can accelerate the fuel corrosion rate. Excessive corrosion can cloud the water, and if through-clad penetration occurs, the concentration of radioisotopes (fission products) on the water can reach unacceptable values. Therefore, in order to maintain the integrity of the fuel for many years, it is necessary to use a purification system, to ensure and maintain good water quality, and minimize corrosion attack of the fuel cladding. As established for the primary cooling system, for storage basins, the utilization of purification systems is based on a filter and resin bed system to keep the water quality within specified limits. Flocculants may be added to remove turbidity in the water. However, the use of flocculants with subsequent filter of the debris would be a special case of water quality management; considerations including impact to criticality and other potential impacts to the safety basis of the facility must be addressed before using any type of flocculants.” (2)

Flocculants are chemicals used in water treatment processes to improve the sedimentation or filter-ability of small particles. Flocculants are used to remove microscopic particles which would otherwise cause water to be cloudy (turbid). Establishing chemistry control of the SFP’s at Fukushima has been very fragmented and has involved minimal attempts at filtration and chemistry addition such as boric acid. Very limited information is available from TEPCO on the total water purity chemistry. It is our assumption that the water chemistry is completely out of specifications mentioned above and has established conditions for optimal corrosion.

“In some cases, mostly in summer, microbiological or algae growth has occurred. Significant biological fouling (biofouling) was experienced in a spent fuel storage pool heat exchanger at one of the nuclear power stations in Canada. The fouling was attributed to a wide variety of bacteria which included pathogenic coli forms. Sulphate reducing bacteria (corrosion bacteria) were either absent or present in low numbers. The action taken to control the bacteria consisted of thorough cleaning of the heat exchanger with a brush, collection of the biological material and incineration of the protective clothing worn by the workers. Appropriate biocides (hydrogen peroxide) at concentrations up to 1000 ppm were added (to the pool water) to control biofouling.” (3)

The normal cleanup systems at the SFP’s at Fukushima have been replaced with at best temporary water system with filtration and reverse osmosis. Approximately 8-9 months after the accident TEPCO did attempt to get the chloride concentration down in specifications after having been in the 6900’s of ppm/ml as compared to a normal level as seen above.

“In Japan at the BWR plants, borated aluminum alloy racks have been used since 1978, and borated stainless steel racks have been used since 1993. In the near future, there is a plan to install borated stainless steel racks in further BWR and PWR plants.” (3)

There is no information that can be gathered to verify if the racks at the Fukushima SFP’s are the original aluminum or the upgraded stainless steel. Knowing the type of rack material is central in determining estimates for corrosion.

“In the USA, one significant event that occurred during handling operations at a spent fuel storage pool involved separation of the top end fitting (nozzle) from the remainder of a PWR fuel assembly as the assembly was being lifted out of a storage rack in the pool at the Prairie Island NPP. There were no radioactive releases from this Westinghouse assembly, and no fuel rod damage occurred. The fuel assembly was subsequently lifted and inserted into a storage position. Intergranular stress corrosion cracking of the Type 304 SS sleeves, which were welded to the top nozzle and mechanically joined to the Zircaloy control rod guide thimbles, was identified by Westinghouse Electric Corporation as the cause of the failure. The Zircaloy guide thimbles appear to have remained intact.” (3)

The only fuel that has been removed to date were 2 unused fuel elements that were removed from unit 4 SFP but since they were relatively new had no major signs of extensive corrosion but there was a lot of debris within the fuel element.

There are some seldom mentioned phenomena that occurs in operating reactor fuel elements which depend on a term called “FUEL BURNUP”. All nuclear power plants (NPP’s) must determine to what degree they allow the reactor fuel element to burn up i.e. what percentage of the actual fuel (like U235) is acceptable to burn up before the fuel element reaches its end of life. Since NPPs are in the business to make profits, many utilities have increased the burn up limits to decrease operating costs.

“To minimize operating costs and increase cost competitiveness, utilities are changing fuel designs and reactor operating conditions, including higher fuel burnups, longer cycles, increased enrichment and new water chemistry. This increase in ‘fuel duty’ and these environmental changes directly impact fuel performance, and new performance issues have appeared. Fuel cladding is a key barrier in containing fission products and it is essential that this barrier is robust and remains intact. Fuel failure occurs when this barrier is degraded and breached. It contributes to increasing plant background radiation, which impacts planned outages and increases workers’ exposure. It can also contribute to the release of radioactive fission products to the environment. Thus, fuel performance should be sufficient to limit radiological releases into the environment and to be able to cope with ALARA issues. Finally, reactor fuel failure does not create confidence in the nuclear power industry and can influence public acceptance of nuclear power generation. For all of these reasons, it is a general goal of modern nuclear utilities to operate with a core free of defects” (4)

When the percentage of fuel burnup is increased many factors are introduced to the fuel elements such as:

• Increased internal pressures within the fuel pellets

• Cracking initiated at a massive hydride layer at the clad outer surface and propagation through the whole cladding thickness (delayed hydride cracking or DHC)
• Insufficient fuel rod support in the assembly due to improper design and/or fabrication, fuel rod vibration due to fluid elastic instability caused by crossflow in the assembly, and flow induced assembly and rod vibration. Debris fretting continues to be a common mechanism for fuel failure in all types of power reactors.

• Pellet–cladding interaction (PCI) fuel failure end

• Plug defects, and several types of end plug weld deficiencies.

These are a few of the major issues that can arise with NPP fuel elements and each can lead to what is called a “LEAKER FUEL ELEMENT” ie: a fuel element that has had some minute cracking or failure of the cladding around the fuel pellet which results in fission products to be dispersed in the primary coolant system. When the leaker fuel element is then placed in a spent fuel pool that same leak may seal up or it may open and release fission products directly into the spent fuel pool. The link above has a detailed section on fuel element leakage in operating reactors as well as spent fuel pool. The main result of a leaker element is the rise in gross activity levels, interaction with the leaker site and the water in the reactor or spent fuel pool. These leaked fission products can result in levels of activity which would not allow work to be done in the area and in the case of Fukushima, would be directly released into the atmosphere. Increased levels of Xe133 is the main isotope that is watched for spotting a leaker. Iodines are also monitored for changes especially when the reactor is shutdown. The most common ratios utilized as indicators of fuel failure are: 133Xe to 135Xe, 133Xe to 138Xe or 85mKr to 87Kr. A significant change in the value of such ratios is a clear indication of fuel failure. In addition, the presence of transuranic isotopes (actinides) in the primary coolant is an indication of the presence of fissile materials in the primary coolant due to erosion of fuel pellets through large defects.

The most significant actinides are:
• Neptunium 239Np;
• Plutonium 238Pu, 239Pu, 240Pu, 241Pu;
• Americium 241Am;
• Curium 242Cm, 243Cm, 244Cm.

A technique called “sipping” has been successfully used to identify potential leakers. (4) Sipping is the most common technique used to locate fuel failures in both PWRs and BWRs. Identification of fuel rod failure is based on the detection of fission product activity released through defects during sipping. The more common radioisotopes measured are xenon and krypton, and cesium or iodine in water samples. Various versions of sipping have been used to detect leaking fuel assemblies.

To date TEPCO has not mentioned the use of sipping in any of the SFPs. No reported efforts have been reported by TEPCO on any efforts to directly determine if fuel element failures, including fuel leakers, have occurred in the pools. Failure to implement “sipping” or equivalent techniques undermines the ability to be proactive and look for changes in the pools which would lead to fission product leakage/release. In addition, TEPCO has not released any information on the number of known defective fuel elements which may be stored in the SFPs including ones that are in canned storage.

Every analysis that has been researched concerning fuel degradation in spent fuel pools has included a qualifying statement, “assuming the chemistry of the spent fuel water is maintained” Clearly the SFPs at Fukushima have undergone unprecedented severe accidents, extreme thermal cycling, unknown chemistry, fires, damage from debris falling into the SFPs including a huge roof truss section which fell directly into the #3 SFP while removing debris. The key and most important parameters went to extreme levels out of specification and were unchecked for months. From these conditions as well as missed attempts to try and bring the water into some form of gross control, the assumptions for degradation rates will be made using the worst case conditions.

SUMMARY

The single most important event in the spent fuel pools at the Fukushima plants is creating conditions in which the fission product contents of a single or multiple fuel element pellets is released into the water and ultimately to the atmosphere. This report has tried to discuss some of the modes of failures that can present during storage in the spent fuel pools. Questions have been asked such as:

• What is the corrosion rate for the metals in the pools and fuel elements

• How will cooling of the fuel elements be maintained to ensure they don’t overheat

• What has all the debris/crud that has fallen and collected in the spent fuel pools do to challenge chemistry control

• What should be done to reduce the impacts of chemistry on the existing stored fuel elements

• What would the conditions be like around the spent fuel pools if the fission products were released to the atmosphere

• Would the activity levels be too high to work in the vicinity of the spent fuel pools if the fuel fission products were released
These are important questions that involve very complicated analysis to predict in a way that can relate to time, such that projections for failures can be made. The idea is to eventually remove the fuel from the spent fuel pools. However, the planning how to achieve this goal is in its infancy, since there has been no case like Fukushima from which to look back and see what was done.

**OBSERVATIONS AND PROJECTIONS**

**SPENT FUEL POOLS**

- BWR fuel elements are more prone to corrosion than PWR elements
- SPENT FUEL pools have undergone extreme conditions as noted earlier
- Specific water chemistry is unknown other than what few samples have been taken focusing on activity levels and isotopes (specifically Cs137)
- SPENT FUEL pools materials of concern are stainless steel, Inconel, some carbon steel and aluminum
- LWR fuel structural material consists mainly of Zircaloy and stainless steel. Zircaloy has proven to be insensitive to any kind of corrosion phenomena (uniform corrosion, stress corrosion cracking (SCC), electro corrosion) in the temperature range <=60°C (3)
- In such cases where Zircaloy and SS are in direct contact, in-service passivation prevents any electro-corrosive phenomena. SS is also resistant to any kind of corrosion. However, some attention has to be given to SCC in the neighborhood of welds. Water chemistry quality control has proven to be a reliable remedy against SCC of SS components (3)
- During underwater storage of LWR fuel, sleeve corrosion has been reported for some LWR fuel with Zircaloy guide tubes and stainless steel upper sleeves. The corrosion phenomena, probably induced at the reactor, have occurred for high carbon content in the upper sleeve material and can jeopardize future spent fuel handling operations. (3)
- The corrosion rates of stainless steels stored for 18-month in pool are very small (10^5 - 10^-4 mm/year) regardless of stainless steel types and pre-treatment histories (3)
- Maximum corrosion occurs at the initial storage stage in small water volumes (cans) at the points of Zr/SS contact (under spacers). Corrosion of SS pool components amounts to 1 µ/year. (3)
- A technique has been developed for assessing the fuel element integrity by calculating the ratio of dissolved '37Cs and '34Cs in water. (3)
- Accelerated concentrations of Chlorides such as was experienced at Fukushima could elevate corrosion by a factor of 100. (3)

**PIPING**

The reference link (5) is an excellent history of some of the erosion/corrosion problems experienced on reactors in Japan

- When predicting rates of erosion/corrosion, pipe sections of non-standard geometry tend degrade faster than the rest of the pipe. Certain parts of the pipe will be subject to differing conditions (due to stream turbulence). These conditions can include temperature, chemical species, and fluid flow rate. As the rate of reduction in pipe thickness is determined by process conditions and materials, predictions cannot be made based on common corrosion data. The same is true of stress corrosion
- Pipe wall thickness reduction is mostly found in the cooling circuit of BWRs. These pipes are made of carbon steel and hence management of water quality (temperature, dissolved oxygen, pH, etc.) as well as material properties (low alloy steel, Austenite stainless steel, etc.) is important
- In general, acceleration of corrosion is caused by two factors: water impurities (sea water etc.) and mechanical stress. Furthermore, the rate of thickness reduction caused by erosion/corrosion is much higher than that caused by corrosion only
- Erosion/corrosion causes are discussed below

- Position (T-junction, curve, joint, and so on, downstream of an orifice etc.)
- Downstream events (water injection, collision, hot water flushing)
- Design (inappropriate structure, unsuitable materials)
- Inappropriate structure composite, e.g. T-junction and water injection
- Fluid-position composite, e.g. hot water flushing through an orifice
- Unsuitable structure and unsuitable fluid complex, e.g. water injection and joint

The linked study arrived at the following conclusions regarding the rate of thickness reduction due to erosion/corrosion

- In general, corrosion proceeds at a rate of 0.15-0.3mm/year
Accelerated corrosion occurs at a rate of 0.3-0.5mm/year

In general, erosion/corrosion proceeds at a rate of 0.5-1.0mm/year

Acceleration of erosion/corrosion occurs at a rate of 4mm/year

The table below lists estimates of corrosion rates for the feedwater pipes at Fukushima. The feedwater pipes are currently being used to supply water to the reactor vessels through a connection inside the reactor building, outside of containment. These pipes were already considerably old, records do not show TEPCO as having replaced these pipes. Browns Ferry NPP was used to estimate feedwater pipe diameter and schedule. (7) According to the nominal pipe thickness of new pipes and the combined corrosion rates, the piping at Fukushima has an estimated life before failure of 3-6 years. TEPCO states they need to continue water cooling for 10 years. Details of the Browns Ferry feedwater system can be found in the companion report "Corrosion At Fukushima Daiichi Explained"

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<th>Thickness (T nominal)</th>
<th>Thickness in MSH 80</th>
<th>Minimum wall thickness (97.5% of nominal)</th>
<th>Lifetime based on accelerated corrosion (mm/yr)</th>
<th>Lifetime based on corrosion / corrosion rate of 4mm/yr</th>
<th>Lifetime based on accelerated corrosion / corrosion rate of 4mm/yr</th>
<th>Estimated life based on loss of wall thickness due to age of piping</th>
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REFERENCES:

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   [link]

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    [link]

   (Companion Paper)
   Corrosion At Fukushima Daiichi Explained
   by The SimplyInfo.org Research Team

   A very critical factor in the aftermath of the nuclear meltdowns at Fukushima is stress and corrosion on metal parts of the reactors themselves and also the spent fuel and fuel pools. In this paper we look at corrosion factors and also the actual pipes in use at Daiichi for the ongoing watering effort.

   Corrosion
   
   The type of metal involved, mechanical loads and the type of fluid in contact with the metal will all impact the corrosion and destruction of pipes and metal. Mechanical loads can include movement of pipes and structures during the initial earthquake, ongoing aftershocks and also movement as equipment broke or shook during the meltdowns. Fluid includes the water that has been sent through the reactor pipe systems, reactor vessels and then flows throughout the plant structures. This also includes the spent fuel pools and the water used to shield and cool the spent fuel. (5)

   ![Diagram](diagram.png)

   Figure 1. Factors influencing corrosion fatigue.
Types of corrosion:

Water Chemistry:

Water and the chemistry of the water plays a considerable role in corrosion. The chemistry of the water is determined by a set of parameters.

**pH:** The reason for controlling pH in the reactor coolant system is to minimize and control corrosion. In reactor facilities (except those containing aluminum components), acidic conditions are detrimental to the nuclear plant materials in a number of ways. An acidic condition in the primary coolant results in processes that are potentially harmful to the system as follows. First, a low pH promotes rapid corrosion by deteriorating or “stripping off” the protective corrosion film, and second, corrosion products such as ferrous oxide (FeO₂) aka rust, which is predominant in the corrosion film, are highly soluble in an acidic solution.

**Oxygen:** Control of the dissolved oxygen content in the reactor facility system is of paramount importance because of its contribution to increased corrosion. Corrosion is dependent on both the concentration of oxygen and temperature.

**Hydrogen:** Hydrogen gas (H₂) and hydrazine (N₂H₄) are the scavenging agents normally used to eliminate dissolved oxygen from the reactor coolant systems. Because hydrazine decomposes rapidly at temperatures above about 200°F (forming NH₃, H₂, and N₂), hydrogen gas is used as the scavenging agent during hot operation and hydrazine is used when the reactor coolant system is cooled below 200°F.

**Total Gas Content:** Total gas is the sum of all gases contained in the coolant system and is made up primarily of hydrogen (H₂), nitrogen (N₂), argon (Ar), and oxygen (O₂). The small amount of two fission gases (Kr and Xe) normally present in the system may also contribute to the total gas concentration.

**Conductivity:** Conductivity of reactor facility water is measured to provide an indication of dissolved ionic substances in the coolant. Reactor coolant conductivity is normally controlled at a level as low as practicable and consistent with pH. Excessively high conductivity levels are an indication of the presence of undesired ions. This condition warrants further investigation to locate the source of the impurity because, in addition to other chemistry problems, it contributes to general corrosion by increasing the reaction rates of the electrochemical cells.

**Chlorides:** The reason for maintaining the chloride ion concentration (salt) at a minimum level practicable is that several forms of corrosion are affected by the chloride ion, and the type of greatest concern is chloride stress corrosion. When high levels of Cl⁻ are suspected, or detected, immediate steps must be taken to eliminate the source and remove Cl⁻ from the system because of the potential consequences. Replacing the water with new clean water is one method as is ion exchange filtering. The method is used to control Cl⁻ concentrations in the reactor coolant by routing water through the ion exchange system. At Fukushima an improvised version of both is being attempted to improve the water quality.

**Fluorine:** High levels of Fluorine are potentially hazardous for two reasons. First, Fluorine promotes corrosion of zirconium by a stress corrosion mechanism at the inner surface of the cladding (fluorine can be introduced to this region because of the existence of small defects or “pinholes” that cannot be completely avoided in the fuel cladding). Second, Fluorine, especially when high, is a major contributor to radiation levels of the reactor coolant. Fluorine can enter through impure ammonia or lithium added to the reactor chemistry. Fluorine is removed by the same methods as salt removal.

**Boron:** Boron is used to prevent reactivity or reactions in the reactor and related parts.

**Radioactivity:** Radioactivity of the reactor coolant system is another factor in corrosion. These impacts include increased corrosion, cladding defects, and fuel element failure or fissions. This level in the cooling water can be controlled by ion exchange filtering.

**Plating:** Also of concern are “crud blocks” and plating. Crud blocks are build ups of sediment that collect in places throughout the cooling system and can cause blockages or adhere to surfaces. Plating is a process where sediments can fuse themselves to surfaces, the interior of pipes or on fuel assemblies. This can block water flow and cooling ability creating hot spots on fuel assemblies.

**Figure 2. Outline of corrosion types.**
TEPCO has set up an improvised watering system at the plant that consists of a network of hoses and pumps to bring water from holding tanks on site. This is pumped into the reactors in an attempt to cool the reactor vessel and the melted fuel though TEPCO is not sure of the actual location of the melted fuel in units 1-3. TEPCO has begun to switch out the hoses for PVC plastic pipe with insulated covers. The plant has had issues over the last winter with freezing and leaking hoses due to the elements.

The water system hooks into the feedwater lines inside the reactor building, these pipes push water to the core spray ring and the feedwater spray ring inside the reactor vessel. TEPCO has had periodic challenges with the watering system losing flow. During one such episode after an earthquake TEPCO assumed debris and crud had broken loose in the pipes causing a partial blockage of the water flow. Clogged pipe problems have presented challenges at the plant as far back as July 2011.

This diagram from Browns Ferry NPP in the US provides a good comparison to the feedwater system in the reactors at Fukushima. It shows the remaining reactor pipes that are still in use at Fukushima Daiichi NPP. The reactor vessel sits in the middle of this pipe array. (2)

Inside the reactor the feedwater spray ring and core spray ring
The pipes for the feedwater system range between 12 to 24 inch outer diameter. These feedwater pipes along with the two spray systems inside the reactor make up the existing reactor systems that need to remain functional for the years TEPCO needs to cool the reactor and melted fuel. TEPCO's most current roadmap shows they need to cool the reactors for the next 10 years. (3)

Summary

Appendix E
(Page 2 of 3)
Carbon Steel / Treated Water

<table>
<thead>
<tr>
<th>System</th>
<th>Drawing Ref</th>
<th>Pipe Size OD (in)</th>
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TEPCO faces a major challenge to keep these existing pipes clear and working for the next 10 years. We did not find any records of these pipes being replaced at units 1-3 at Fukushima Daiichi NPP. The feedwater and core spray rings inside the reactors were replaced as part of the annulus replacement work that was done at units 1-3 between 1997 and 2001. The pipes are about 42 years old and have suffered the extreme conditions of the triple meltdown accident. TEPCO has not proposed any plan for dealing with this issue.

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   R.-P. Berg - Bundesamt für Strahlenschutz, Salzgitter, Germany
   R&RATA # 4 (Vol.2) 2009, December

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