

Historical and Present Issues on Secondary System Chemical Treatment and Corrosion in PWR/VVER Units; including Materials Behaviour

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A handwritten signature in black ink, appearing to read 'P. Rudling', is positioned above the name of the signatory.

Mr Peter Rudling, President of ANT International

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List of Abbreviations

Unit conversion

1 Introduction

This report describes and explains the past and present issues related to secondary system chemistry and materials behaviour.

It starts with the relation between design and material evolution influence on chemistry selection, explaining in the past why plants switched from phosphate to All Volatile Treatment (AVT), stringent chemistry guidelines and, as a consequence, the use in several countries of condensate polishing plants in addition to tight condenser design (both at water box and with more resistant tubing materials (titanium and stainless steel instead of copper).

The evolution of Steam Generator (SG) design for blowdown and Tube Support Plate (TSP) is then explained in relation with encountered types of SG tubing corrosion.

Then the evolution of the very important SG tubing material (Alloys 600 MA, 600 TT, 800, 690 TT, and 18-10 of VVER) is largely detailed. The link with various degradations (wastage, denting, Intergranular Attack/Stress Corrosion Cracking) is explained.

The switching from ammonia to amine treatment or AVT at high pH (H-AVT) is discussed in relation with Flow Accelerated Corrosion (FAC) of carbon steel. Corrosion products transport mitigation is explained as well as the optimum hydrazine added concentration for mitigating oxidising environment in the SG.

The reagent selection is detailed, based on design and material evolution, on encountered degradations and on new issues. The advantages and inconveniences of H-AVT and various amines treatments are explained. The optimum hydrazine concentration depending on various parameters is described. The distribution of the reagent within the secondary system is covered as well as the behaviour of various impurities (Na, Cl, organic acids, etc.).

The important issue of growing importance, corrosion products generation, deposition and elimination, is fully covered with the selection of adequate treatment, while other remedies are briefly listed (dispersant addition, film forming amine, chemical cleaning, sludge lancing etc.).

New challenges with costs and environment (liquid and solid wastes) in addition to safety, primary coolant integrity, long term behaviour of secondary system materials are explained. This covers the best selection of reagents and purification system (limitation of the use of condensate polishing plant, operating mode of steam generator blowdown).

The report clarifies the potential influence of lead on various steam generator tubing materials, and different chemical environments.

The integrity and long-term behaviour of the plant are largely considered.

The knowledge used to write this report includes a worldwide power plant experience feedback with different designs and chemistry options, vendors and large organisms criteria, and also R&D results.

2 Design and Materials Evolution, Associated Chemistry evolution

2.1 Condensers – Phosphate treatment, Wastage

Up to 1974, the various vendors were recommending the application of phosphate treatment in the steam generator (SG), which was applied in almost all the plants in operation.

The purpose of this old treatment based on phosphate addition (PO_4) with a specific ratio of Na/PO_4 was to buffer impurities entering the SG from condenser leaks. This means that whatever the type of cooling water or other impurity ingress, either alkaline or acidic forming, the pH in the SG was remaining almost constant due to the presence of sufficient sodium phosphate amount in the SG. The most frequent impurities ingress was due to leaks from condenser with copper alloy that was the common selected material due to its property of high thermal conductivity.

The various advantages and inconveniencies of actual materials that may be selected for condenser tubing is explained on Figure 2-1. The good conductivity of copper alloy is compensated on titanium tubing by a lower tube thickness, which is acceptable due to its good resistance to corrosion. For stainless steel, 304L has been applied but suffered from corrosion in some cases, depending on the type of cooling water; consequently, 316L is a better selection if stainless steel is selected instead of titanium for economical reason. Stainless steel has been chosen instead of titanium when the price of this metal drastically increased.

For units cooled by sea water, where only extremely low leaks are acceptable, titanium tubing should be considered as mandatory, whatever the price of this metal, for avoiding the risk of small leaks that are not compatible with specifications at SG blowdown.

Although condensate polishing plant may be considered as an option to eliminate the impurities from such leaks, this is not a clever choice due to all its disadvantages that are explained in section 5.2. Then, even if condensate polishing plant is in service, the condenser leak will have to be eliminated soon or later. Finally, it is much better to avoid the ingress of impurities through the selection of a tight condenser than trying to eliminate the pollutants once in the secondary system.



Material	Copper alloy	Stainless steel or Titanium
Advantages 	<ul style="list-style-type: none"> ■ Lower cost ■ Bacteriostatic → No need of biocide treatment with associated wastes 	<ul style="list-style-type: none"> ■ Good behavior to corrosion (mainly Ti or 316L) ■ Few leaks, especially for Ti ■ No undesirable chemical releases
Inconveniencies 	<ul style="list-style-type: none"> ■ Degradation (abrasion) ■ Life duration, leaks ■ Release of Cu, Zn ■ Low pH in secondary system 	<ul style="list-style-type: none"> ■ Development of biofilm ■ Potential need of biocide treatment for NPP cooled by river, with associated wastes ■ Increased risk of fouling

Figure 2-1: Evolution and rationale for condenser tubing selection.

However, with the new generation of PWR units of higher power and heat flux, it appeared that locally either acidic or alkaline environment occurred leading to SG tubing corrosion as explained below in this section 2.1.

Consequently, American vendors and French licensee urged their utilities to switch from phosphate to All Volatile Treatment (AVT).

Due to the absence of buffering reagent in the SG under AVT chemistry, it was mandatory to drastically decrease the allowed concentration of impurities in the SG that were able to concentrate and easily develop acidity or alkalinity, and thus SG tubing corrosion.

At first, utilities considered the new specifications from the vendors hardly applicable, but with better condenser technologies and/or addition of condensate polishers, this appeared possible.

The main evolution in condenser design consisted in selecting more resistant tubing, particularly titanium for plants cooled by sea water where only extremely low levels of leaks were acceptable with the new specification, due to the high concentration of impurities in cooling water as compared to river or lake water.

In addition to the better resistance to corrosion of titanium as compared to copper alloy, this new material has the great advantage of not developing small leaks that cannot be easily localised as it is very possible with copper alloys.

Another improvement in condenser design consisted in specific water box design (double tubesheet (TS) or various design to avoid ingress of sea water) as shown on Figure 2-2.

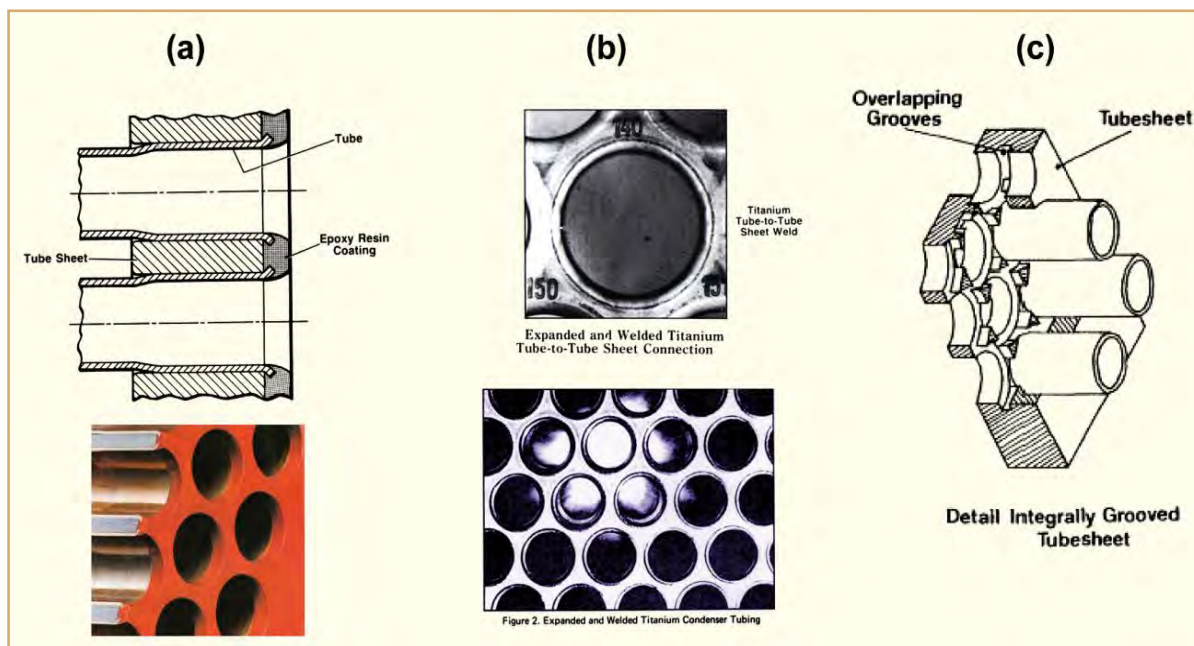


Figure 2-2: Condenser tube to tube-sheet connections: (a): tubes rolled and sealed by epoxy resin coating; (b) Tubes rolled and welded into tube-sheet (Leak tight condenser designed by Siemens-KWU); (c): Integrally grooved double tube-sheet technology [Zoerner & Termuellen, 1985].

For plants cooled by fresh water (river, lake), Utilities selected either titanium or cheaper tubing such as stainless steel or in some other cases, kept copper alloys, in most cases with condensate polishing plants for eliminating small or moderate condenser leaks.

Consequently, part of the chemistry evolution is linked to condenser technology tightness evolution, but also to power increase and to alloy type of SG tubing.

Phosphate treatment and Wastage

The Phosphate chemistry was well known, having been used in the fossil fired power plants to increase the pH value in the boiler water and to provide chemical buffer to prevent pH excursions, which might be caused by chemical reactions and/or hydrolysis of the concentrated impurities at high operating temperatures.

The pH increase by PO_4 s is based on its reaction with water, which produces sodium hydroxide:



After the first degradations of alkaline Stress Corrosion Cracking (SCC) on the secondary side of SG tubes (ODSCC) with Alloy 600 observed on recent units of higher power in the early 70's, a lower Na/PO_4 ratio has been selected. But it induced acidic corrosion, called wastage, that is tube thinning beneath the corrosion product deposits, as illustrated on Figure 2-3. Several Na/PO_4 ratios with a narrow range have been selected to try keeping the chemistry in a more controlled chemical environment. However, due to local concentration of reagent and impurities, it appeared impossible to avoid corrosion: Stress Corrosion Cracking (SCC) under alkaline environment or wastage under acidic environment, explaining the decision of American vendors to move to AVT in 1974.

In the second part of the 70's, KWU decided to remain on PO_4 treatment since SG tubing Alloy 800 was not sensitive to IGA/SCC as Alloy 600, but later on finally also moved to AVT due to unavoidable wastage.

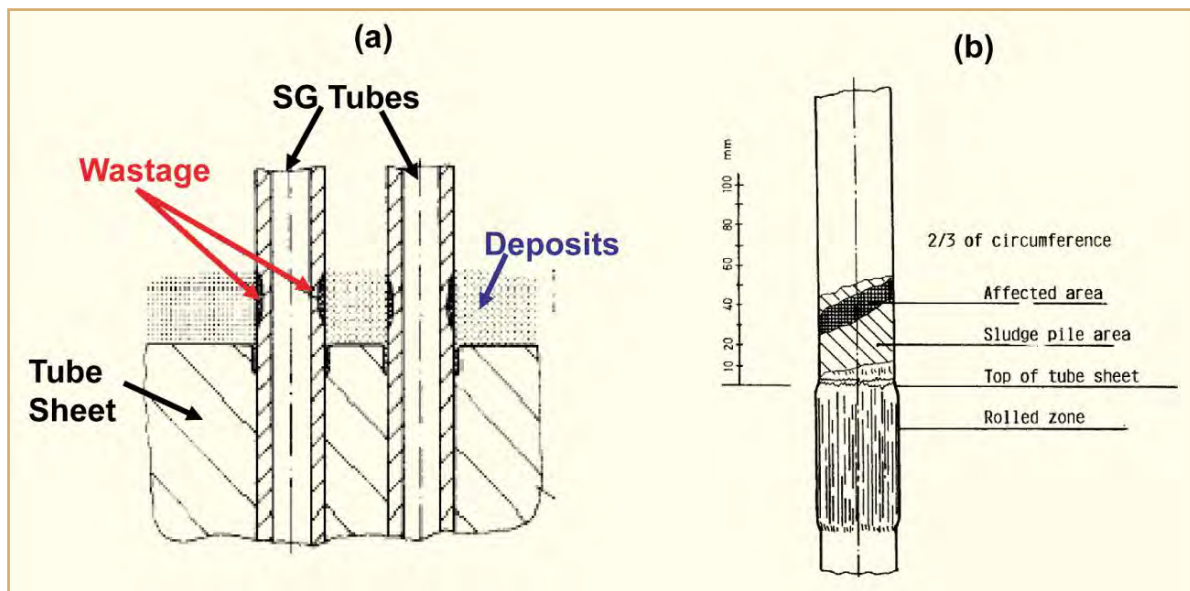


Figure 2-3: (a) Schematic picture of wastage corrosion mechanism [Schroeder, 1984]. (b): Pulled tube with wastage corrosion [Odar, 1987].

In the second part of the 70's, when US vendors asked to their utilities to apply AVT, German vendor Siemens/KWU decided to remain on PO_4 treatment since SG tubing Alloy 800 was not sensitive to IGA/SCC as Alloy 600, claiming this treatment was satisfactory for the German design. Later on, in the 1980's, with wastage occurring in units of German design, after a number of years ranging from 8 to 2 years, for temperature increasing from 255°C to 280°C, KWU finally also specified AVT chemistry.

2.2 Lower specification limits

The application of AVT chemistry without buffering effect induced more stringent specification, with, in 1974, limits that were more or less corresponding to the concentration of impurities that was achievable at SG blowdown. These lowest concentrations were depending either on existing technologies of the lowest leak rate that could be localised in the condenser, or by addition of condensate polishing plant to eliminate the soluble impurities entering from condenser leak.

Almost immediately after switching from phosphate to AVT treatment, several units suffered from denting at Tube Support Plate (TSP) level. This was due by corrosion of TSP made of carbon steel that were corroded mostly with impurities coming from seawater leaks in condenser that were developing acidity in the SG. Such corrosion was even more severe in presence of oxygen or oxidising corrosion products that were common at the time with high level of oxygen entering the secondary system from the condenser. The denting mechanism is

illustrated on Figure 2-4 for TSP level and Figure 2-5 at top of tubesheet. The locations are different but the mechanism is the same: a rapid corrosion of carbon steel, which expands when it oxidises, since the density of the oxide is about twice lower (4) than the density of the parent metal (8). As a result, the oxides squeeze the SG tube that cracks due to the very high stress level.

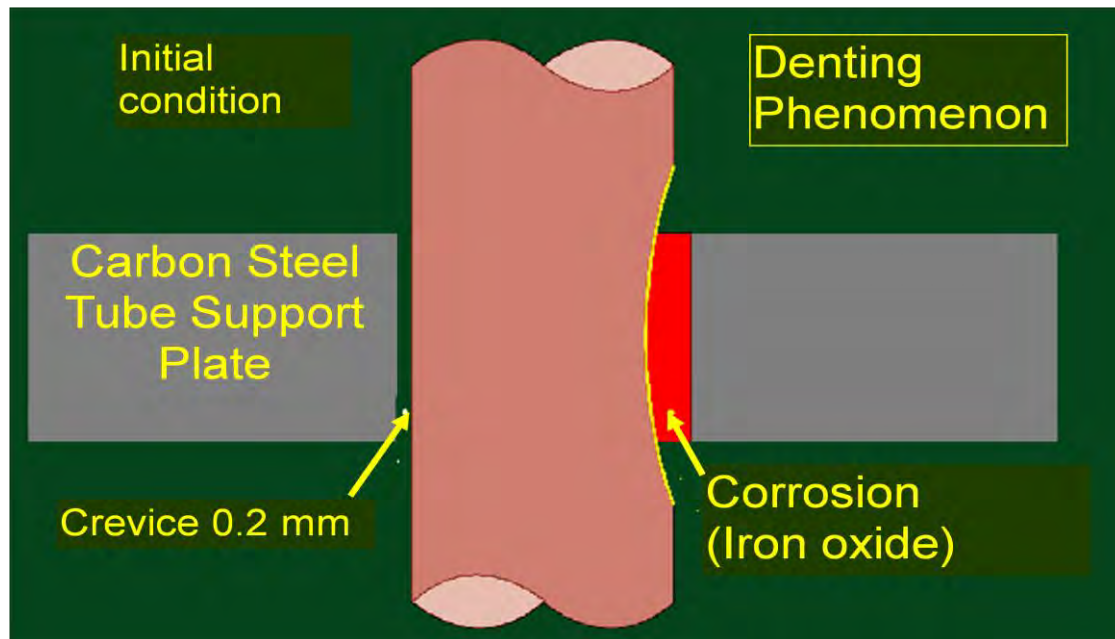


Figure 2-4: Simplified Principle of denting at TSP made of carbon steel [Nordmann et al., 1982].

The chemistry remedial action against denting has been to minimise the sea water ingress (that was inducing acidity and the oxygen level in the condenser that induced the transport of oxidising corrosion products in the SG as well as allowing an oxidising environment, detrimental for CS corrosion.

Then, it had been noticed that, when primary to secondary leaks were occurring, the presence of boric acid in the secondary system was able to decrease the kinetics of denting, as monitored by hydrogen generation ($3 \text{ Fe} + 4 \text{ H}_2\text{O} \rightarrow \text{Fe}_3\text{O}_4 + 4 \text{ H}_2$). A remedy, confirmed by laboratory tests in model boilers, has been to add boric acid in the secondary system of affected units [Nordmann et al., 1982].

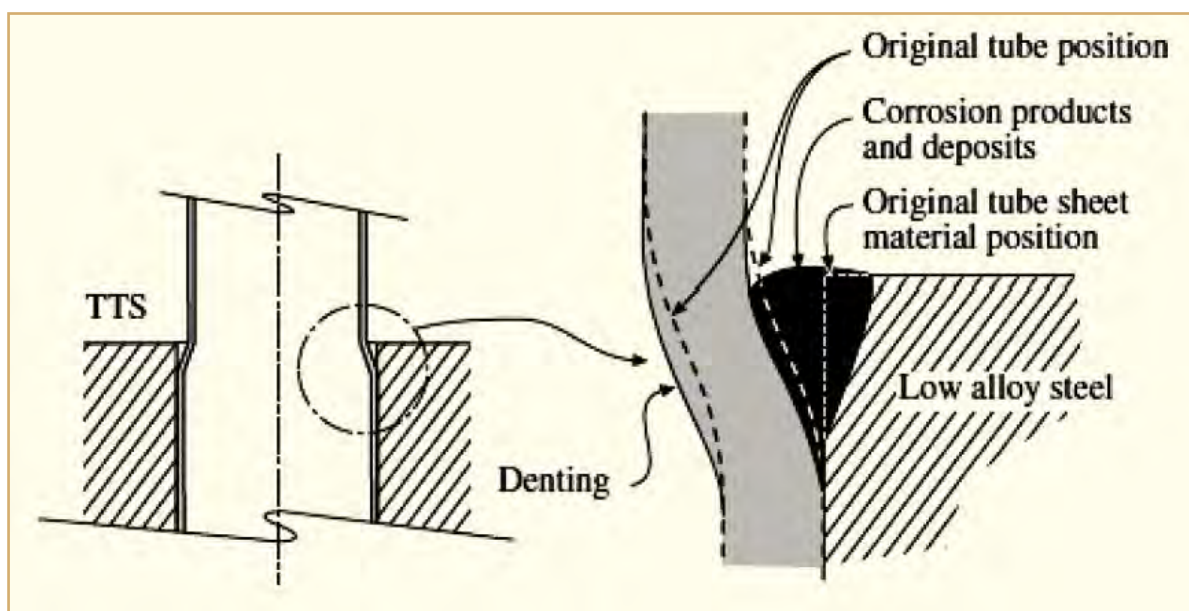


Figure 2-5: Schematic view of denting at the TTS crevices [Staehle & Gorman, 2002].

Besides the application of more restrictive chemistry specifications at SG blowdown, thanks to better elimination of condenser leak, several SG, highly affected by denting, have been replaced with new technologies of TSP made of stainless steel instead of carbon steel, and with broached or egg crates instead of circular drilled holes where the impurities were concentrated and inducing corrosion. Most of seawater cooled units having condenser with copper alloy tubing that were not tight enough replaced their SG. In the same time, more restrictive specifications were imposed.

Although denting problem had been solved, on recent PWR units of high power (≥ 900 MW) and heat flux that were in operation in the 70's, with SG tube made of Alloy 600 MA, highly sensitive to SCC and/or SCC with Intergranular Attack (IGA/SCC), the application of guidelines specified in the mid 70's was unable to avoid SG tube degradation.

Thus, more restrictive levels of impurities (sodium, chloride and sulphate or cation conductivity) have been specified for SG blowdown. However, on most of the units with Alloy 600 MA, whatever the level of impurities at SG blowdown, it appeared almost impossible to completely avoid IGA/SCC that occurred mainly at TSP level and, in a less extent, at the bottom of tube sheet. Particularly in units where the chemistry in the SG was alkaline: units cooled by alkaline forming river water or alkalinity coming from condensate polishing plant throw.

In fact, SCC is the result of a combination of three detrimental parameters: (1) a sensitive material, (2) a high level of stress and (3) a corrosive chemical environment, as illustrated on Figure 2-1. In the case of Primary Water SCC on the primary side of SG tubes, corrosion occurs due to a sensitive material (Alloy 600) in presence of high stress levels (e.g. at tube expansion, small radius U-Bends). In the case of secondary side of SG tubing, SCC occurs mainly in presence of sensitive material (Alloy 600) and detrimental chemical environment (alkaline) and even more in locations with higher level of stress.

An obvious remedy on chemistry was the addition of boric acid in the secondary system, for neutralising the alkalinity. The application of this treatment was easy since it had already been used to mitigate denting.

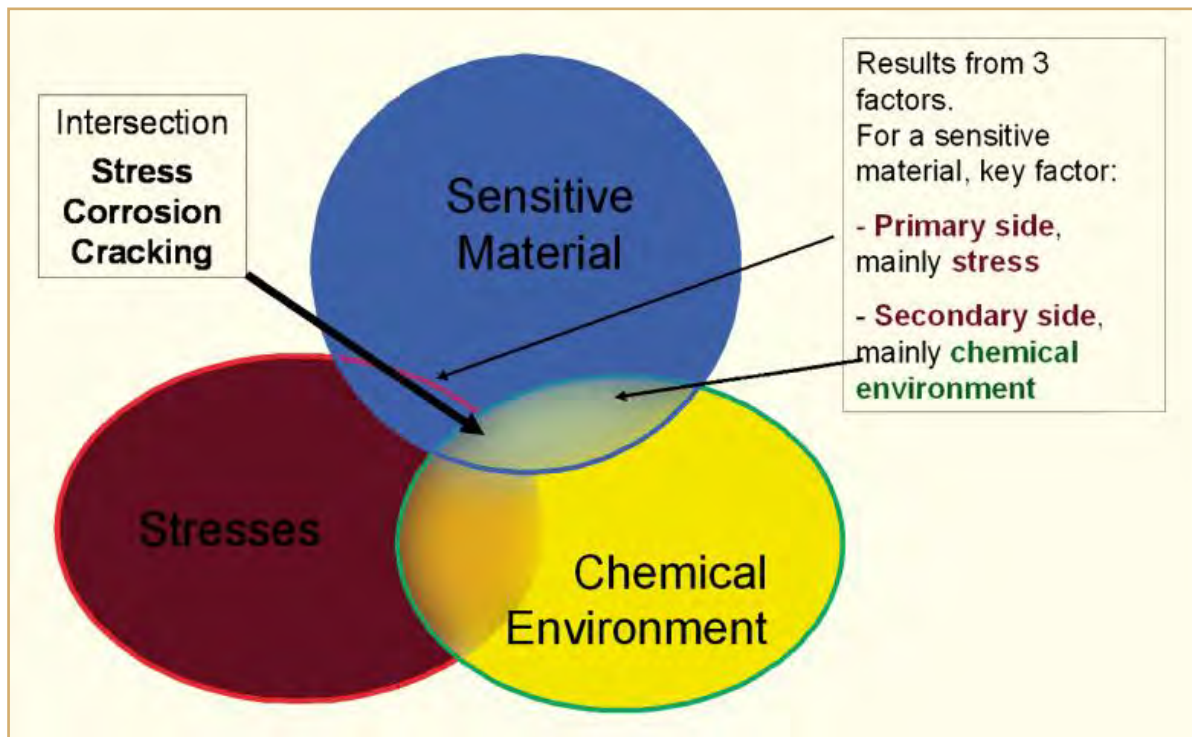


Figure 2-6: Stress Corrosion Cracking influencing parameters.

Within these detrimental parameters for IGA/SCC, Figure 2-7 clearly shows the difference between (left part) pure SCC, where the level of stress contributed to the corrosion, like at TSP level or in the transition zone at the bottom of the tube and (right part) IGA due to dissolution of the grain boundary in a highly alkaline environment. In this case, the chemical environment plays a higher role on corrosion than the stress level.

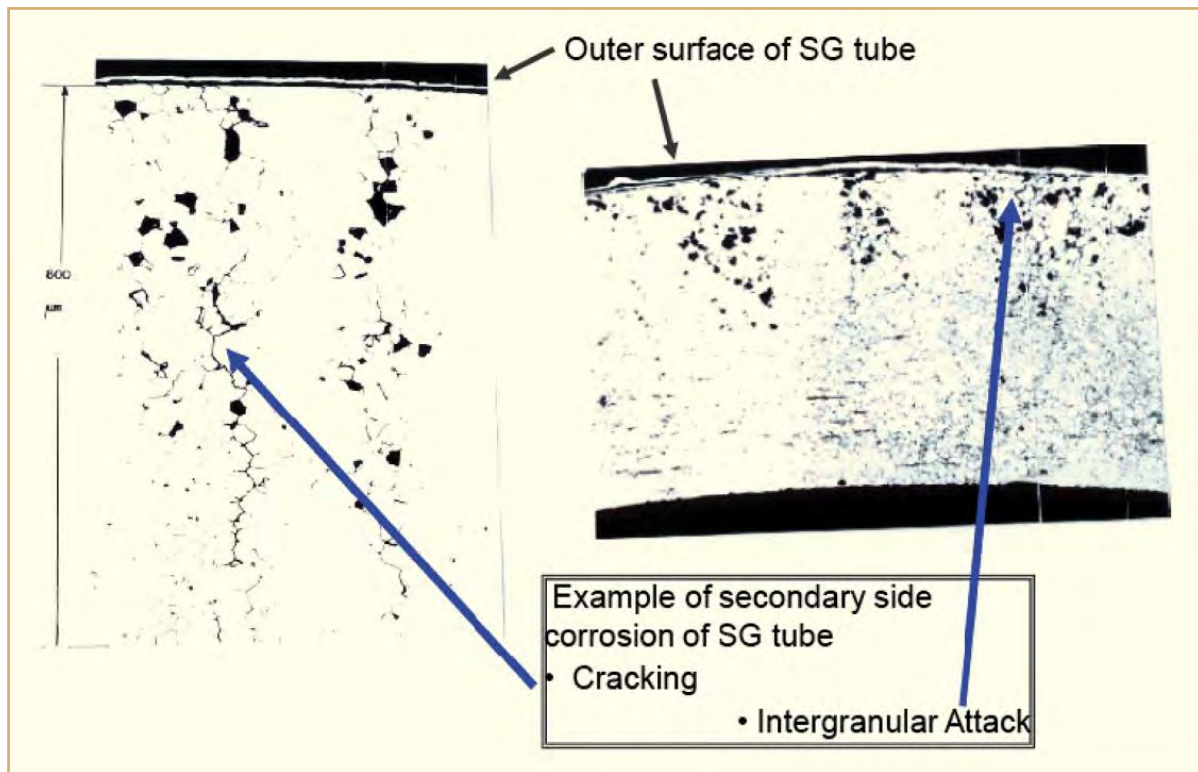


Figure 2-7: Micrograph of Intergranular attack view of stress corrosion mechanism on a Fessenheim 1 SG tube [Odar & Nordmann, 2010].

With the evidence of degradation of Alloy 600 MA tubing, water chemistry guidelines and specifications have continuously been strengthened up to the point where, for some of the guidelines, almost no pollutant ingress is accepted. Such stringent specifications have been applied in countries with Alloy 600 (MA or TT) to all their PWR units because, up to 1989, there was no plant in operation with Alloy 690 TT. Although more resistant SG tubing, mainly Alloy 690, replaced the sensitive Alloy 600MA, once a restrictive specification had been applied, with the demonstration of its feasibility, extremely limited relaxation has been applied for PWRs with Alloy 690.

In addition, some of the specifications focused on the wrong parameters with unnecessary restrictive values on some of them. Indeed, the main cause of SCC was the presence of a highly sensitive material Alloy 600 as explained by Coriou [Coriou et al., 1959], due to a high content of nickel and insufficient content of chromium, Figure 2-8.

This well known figure highlights the following:

- In the early times of nuclear energy, after naval application, the identified risk was mainly sea water ingress into the condenser, with the presence of chloride, which is the most important ion in sea water. Stainless steel being sensitive to corrosion by chloride, American vendors, influenced by INCO (selling nickel) selected material Alloy 600 (also named Inconel 600), with a very high nickel content (72 %) to ensure a good resistance to corrosion by chloride.
- The drawback of such an alloy is that the chromium content is insufficient with the consequence of having a material highly sensitive to SCC, even in absence of any impurity.
- Finally, the use of Alloy 600 instead of stainless steel (304 or 316) has been the biggest mistake in the western design during the 60's and 70's with important consequences of SCC on many components of the reactor coolant system.

3 Reagents selection and Impurities behaviour

3.1 Historical evolution of secondary side treatment

The purpose of this chapter is not reflecting any nostalgia or reporting old stories, but to explain to tell young engineers the reason for treatment evolution and justification for the current various possible choices. This may help them at better deciding what to apply in each case, without having the need to evaluate again what has already been tried.

In the early times of PWR, the main applied treatment was “phosphate” with a defined ratio of Na/PO_4 to ensure a buffering of the water in the SG and avoid an environment too acidic or too alkaline in case of pollution, particularly from condenser leaks. However, with the increase of power of NPP, this phosphate treatment caused wastage, an acidic corrosion described in section 2.1. Then, whatever the selection of the Na/PO_4 ratio, either wastage or alkaline corrosion occurred on Alloy 600, highly sensitive to SCC. The various vendors progressively recommended to switch from phosphate to All Volatile Treatment (AVT) without any addition of a buffering compound.

At the beginning, with the presence of copper alloys in many NPP, a treatment at low $\text{pH}_{25^\circ\text{C}}$ (≈ 9 or 9.2) mainly with ammonia was applied in most PWR and VVER units.

According to the FAC explained in section 2.4, it is now recommended to apply either ammonia at a high $\text{pH}_{25^\circ\text{C}}$ (>9.8) called high AVT (H-AVT) or to select a treatment with amines, allowing a higher pH at temperature (pH_T) even with a low $\text{pH}_{25^\circ\text{C}}$.

The history of secondary side treatment is linked to the encountered corrosion problems (wastage, IGA/SCC on Alloy 600, FAC, corrosion products transport and deposition, releases into the environment, operating costs, etc.). These are shown in Figure 3-1.

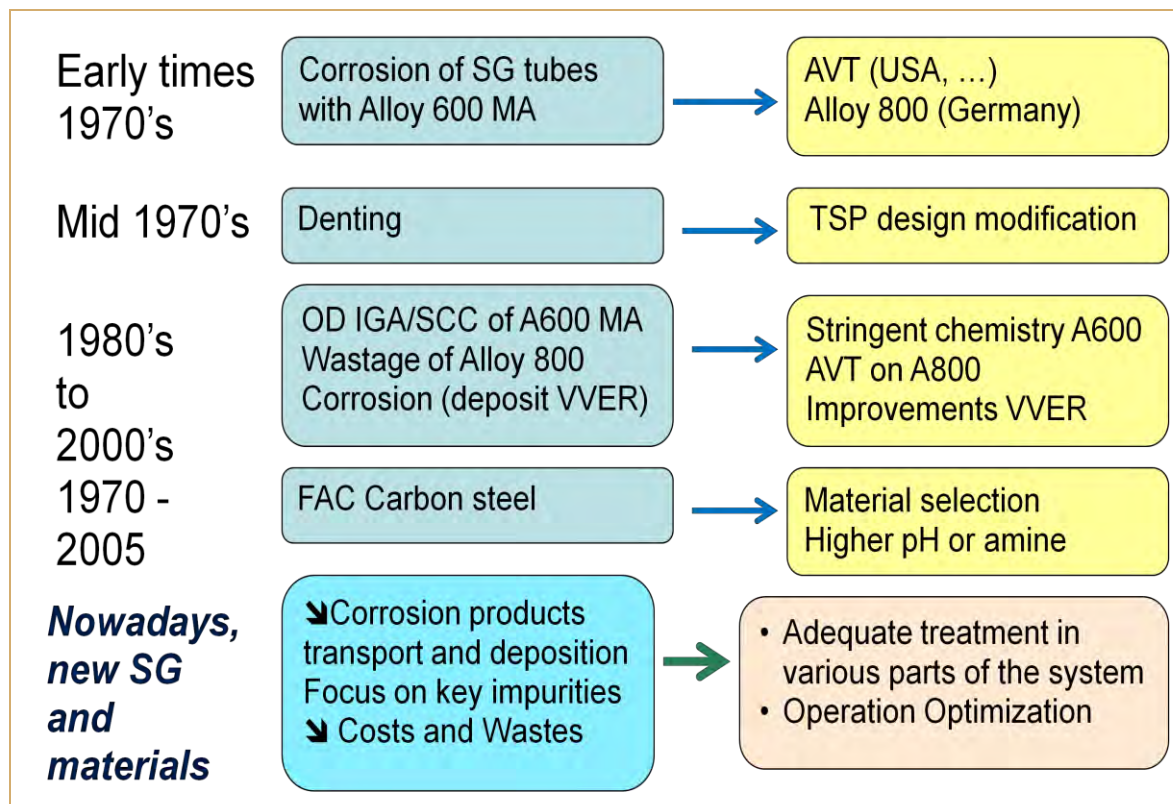


Figure 3-1: Evolution of corrosion problems and treatments in the secondary system of PWR and VVER units.

The evolution from phosphate treatment to the main satisfactory options is listed in Table 3-1 for various countries and in Figure 3-2 for the countries with the largest fleets, except Russia where mainly ammonia has been used for a long time. The main evolutions in various countries are as follows:

- As already explained, phosphate treatment has been replaced by AVT in 1974 in many countries, due to either wastage and/or IGA/SCC, except in plants with German design and Alloy 800 SG tubes, which were not suffering from IGA/SCC. However, German plants also moved to AVT later on (in the 1980's) due to wastage. They selected H-AVT (AVT with ammonia at $\text{pH}_{25^\circ\text{C}} > 9.8$) for FAC mitigation.
- French plants with copper alloy (condenser, LP heaters) switched to morpholine at low $\text{pH}_{25^\circ\text{C}}$ (9.2) to mitigate FAC, since ammonia at such a pH does not protect carbon steel from FAC. Many plants in other countries remained much longer to ammonia treatment at an insufficient pH for a significant time, at least to the 2000's for several countries. This explains for example the severe accident from FAC at Mihama plant in 2004.
- US plants under EPRI recommendation moved to amine treatment much later than the French plants, after having investigated various amines. Ethanolamine (ETA) has been the most largely used amine, starting in the middle of the 1990's.
- Many countries progressively in the 1990's, 2000's and 2010's also switched either to H-AVT or, in an increasing number of cases, to amine treatment, ETA being the most largely used.
- Boric acid treatment has been applied originally to decrease denting at TSP level in some of the plants with the sensitive design of circular holes and carbon steel TSP. Most of the affected units had to replace their SG and stopped boric acid injection. Then, this treatment has been also applied for neutralization of alkalinity to decrease the progression of IGA/SCC in several plants with Alloy 600 MA SG tubes.
- EPRI also considered some other inhibitors, particularly titanium, which has not been largely applied since it was not more efficient than boric acid, even less, for trying to slow down IGA/SCC progression. In addition, EPRI also proposed later on the application of Molar Ratio Control (MRC) for the same purpose. This consists in addition of some NH_4Cl , if necessary, to decrease the Na/Cl ratio < 1 for avoiding an alkaline environment detrimental for IGA/SCC.
- Japanese units, before SG replacements, applied a highly reducing environment with a very high hydrazine concentration, also for trying to find a remedy to mitigate IGA/SCC. However, such a treatment was not really suitable and even more bringing other inconveniences with the risk of reducing sulphate into other sulphur compounds highly corrosive. Then such a high hydrazine being decomposed into ammonia in large amounts, did not allow the possibility of applying an amine treatment that should have been necessary for FAC.
- Most of the other countries, considering that SG with sensitive Alloy 600 MA tubes could not continue their operation for a long time, decided to replace them instead of applying sophisticated chemistries with very limited efficiency.
- In addition to a higher pH_T with either H-AVT or amine treatment, several remedies have also been applied. This is the case of addition of dispersant, applied in a few US plants.

Table 3-1: Evolution of secondary side treatment in various countries.

Option	Reason	USA	France	Germ. ¹	Japan	VVER	Others	Result
Phosphate	Buffer	Yes	N/A	Yes	N/A	?	Y & No	Wastage if Na too low
AVT	Wastage	1974	1977	1980's	Initial	Yes	Most	Denting some design
AVT Boric acid	Denting	Yes	No	No	Some	No	Few	Some efficiency
AVT Boric acid	IGA/SCC	Yes	Few units	No	No	No	Few	Some efficiency
H-AVT	FAC	No	Few units	1980's	Yes	No	Many	Efficient if pH>9.8
Morpholine	FAC	Few units	1984 Most	No	No	Few	Few	Good for FAC Organic acids
Ethanolamine	FAC	Start 90's now 50%	In Late 2010's	No	50%	Few	Many	Replace ammonia or morph. in some NPP
Other amines	FAC	Some	No	No	No	No	No	Mixing, optimisation

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Some NPP (PWR, VVER) still operate with ammonia at a low pH_{25°C}, mainly for historical reasons, or due to the presence of copper alloys and insufficient knowledge with amine treatment, or to the presence of condensate polishing plant or for any other reason. Such a treatment with only ammonia is considered as inadequate, since it induces an excessive corrosion rate of carbon steel present in the steam-water system. This is why, either H-AVT or amine treatments are discussed below.

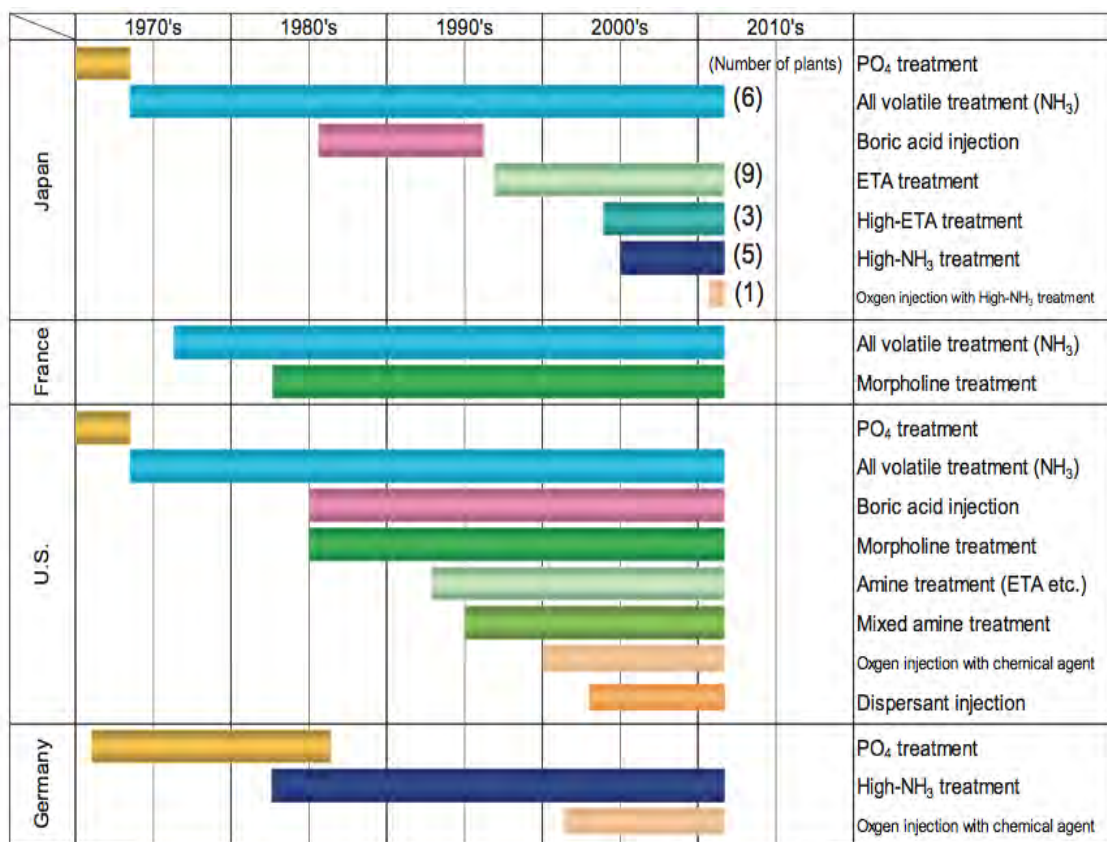


Figure 3-2: Secondary Side Water Chemistry Modifications in some countries.

3.2 H-AVT & Amines treatment

The impact of materials on the pH for the secondary water treatment is summarized below in Figure 3-3.

The key points are the following:

- In presence of copper alloys, it is not possible to increase the pH above 9.3 otherwise the corrosion of the components with copper will be dissolved rather quickly with 2 consequences :
 1. the decrease of integrity of these components, and
 2. undesirable transport of copper compounds to the SG where they may accelerate various types of corrosion, particularly pitting or IGA/SCC.
- In presence of carbon steel, a $pH_{25^{\circ}C}$ in the range 9-9.8 is only acceptable and requires at least the use of amine treatment .
- The economical and environmental impact on Ion Exchange Resins (IER) does not allow to operate at a very high $pH_{25^{\circ}C}$, and in no case > 10 . Below this value, the pH and treatment impact depend on many parameters that are discussed in detail in sections 3.2.1, 3.2.2 and 5.2. The overall principal is to avoid a regeneration or replacement frequency of IER that is too high with high amounts of liquid or solid wastes.
- Stainless steels or titanium do not have any direct impact on the pH and treatment selection.

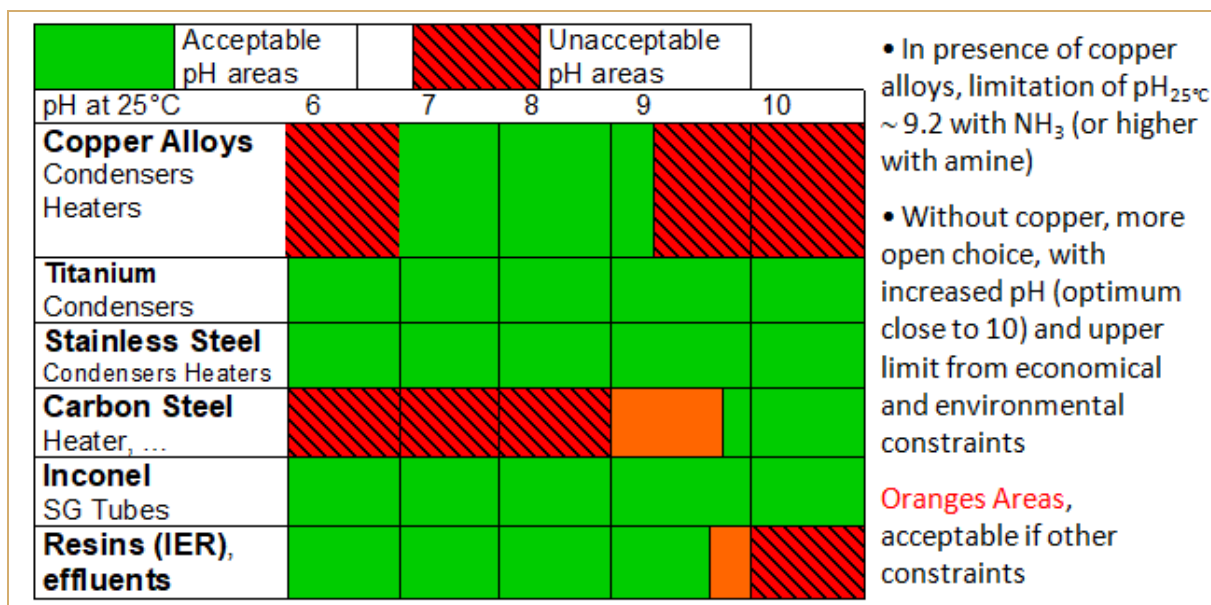


Figure 3-3: Strategy for pH $_{25^{\circ}C}$ in Feedwater.

For the same $pH_{25^{\circ}C}$ imposed by some materials or by other constraints, the pH at operating temperature pH_T in various parts of the steam-water system depends on the reagent, since the evolution of alkalinity versus temperature differs from one reagent to another reagent.

The key point is that, for the same $pH_{25^{\circ}C}$ in feedwater, the pH_T is lower with ammonia as compared to that obtained with amine treatment. This is why, if ammonia treatment is selected, a sufficiently high $pH_{25^{\circ}C}$ is mandatory. This is why, either H-AVT or amine treatments are discussed below.

The objectives of minimising Carbon Steel (CS) corrosion are not only to avoid FAC and deterioration of the CS components, they also aim at minimising the transport of corrosion products into the SG with the many associated inconveniences.

3.2.1 H-AVT

Within the two main options of steam-water treatment in PWR or VVER units, a simple one is the H-AVT with a $\text{pH}_{25^\circ\text{C}}$ of > 9.8 (9.8-10). Its simplicity is related to 2 key points:

- Only one reagent for getting the target pH value instead of at least two with amine treatment where anyhow, ammonia is produced from hydrazine decomposition. Consequently, less measurements of different reagents for pH adjustments.
- Potential addition of only hydrazine if ammonia is not too much eliminated. Less reagents to dose in the system.

With ammonia, as already explained, a very high FW ammonia concentration (5 to 10 mg/kg), corresponding to pH_{25} 9.8 to 10 is necessary to achieve the threshold of pH_T in any part of the system for protecting the Carbon steel (CS) surfaces. Based on Operating Experience Feedback (OPEX), if CS is used in steam drain parts, instead of low chromium or stainless steel, the FW pH_{25} of 9.8 might even be insufficient to reduce the FW iron concentrations down to about 1 $\mu\text{g/kg}$.

High AVT is not compatible with the use of Condensate Polishing Plants (CPP) since the molar concentration of ammonia is high and the regeneration frequency of IER would be extremely high and unpractical.

In addition, even without CPP, the high concentration of ammonia requires a high frequency of IER regeneration at SG Blowdown (SGBD). Then the operation of cationic resin of SGBD purification system is less efficient for sodium purification if saturated by ammonia as compared to the saturation by morpholine, as explained in section 5.2, due to the difference of affinity of the resin for ammonia and morpholine or in a less extent for ETA. Consequently, cationic IER cannot be operated after saturation by ammonia, if the sodium limit at SGBD is not very low, as it is the case now in most of PWRs.

To achieve High-AVT chemistry, only hydrazine may be injected. Additional ammonia injection is not necessary in some plants like the Siemens-KWU designed plants, because the secondary side is very leak tight. Accordingly, the capacity of the Condenser Air Removal is not designed too large. This results in relatively small ammonia going out of the secondary system. Therefore, the continuous ammonia removal by SGBD and Condenser Air Removal systems can be balanced only by thermal decomposition of the hydrazine excess to achieve the required FW pH_{25} value of ≥ 9.8 . The advantage is the addition of only one reagent and less monitoring. The optimum hydrazine to get a reducing environment is ranging from 50 to 100 $\mu\text{g/kg}$ (ppb).

However, in some units, if this concentration is not sufficient to get the target pH, it is recommended to add some ammonia rather than increasing hydrazine to high values, just for having it thermally decomposed and producing ammonia. The reason is the high cost of hydrazine as compared to ammonia and, moreover, the carcinogenic properties of hydrazine. Thus, only the necessary concentration of hydrazine to be in a reductive environment should be added.

Finally for some vendors or utilities, the advantage of H-AVT versus amine treatment is the absence of organic acids coming from the reagent decomposition. This is important for example for the German vendor of the turbine requiring low values of organic acids. This is also important if the purity of the secondary system is monitored at low value by cation conductivity, since organic acids induce a blank of cation conductivity.

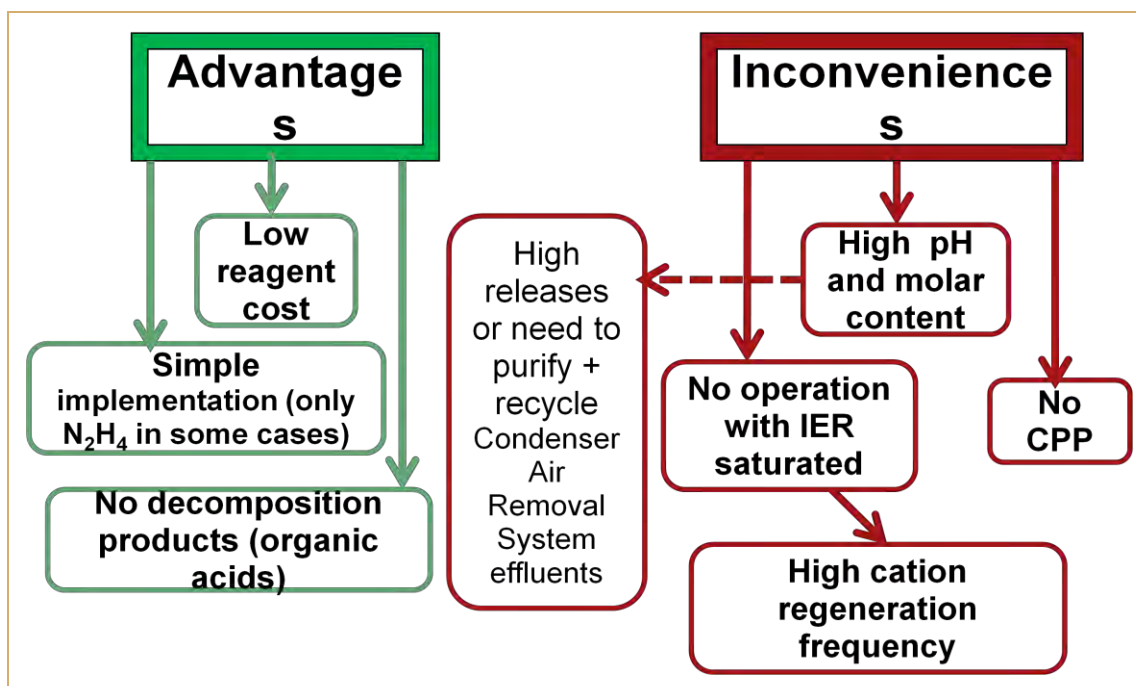


Figure 3-4: Advantages and Inconveniences of Ammonia Treatment at high pH, H-AVT.

3.2.2 Amines

There are some plants where there is the choice between H-AVT and amine treatment and others where amine treatment is the only reliable possibility. This is the case of plants with:

- (1) copper alloys requiring to operate at a feedwater (FW) pH₂₅ <9.3 for which ammonia does not provide a sufficient protection against FAC,
- (2) with CPP in permanent operation for some reason, although this should not be done any more with tight condensers but that is still done to cope with sometimes unnecessarily restrictive EPRI guidelines,
- (3) if cationic IER at SGBD is not regenerated, as it is the case in French PWR units to avoid contamination during regeneration and to minimize potentially radioactive liquid wastes.

Nevertheless, even if in all other cases, H-AVT treatment is feasible, there is a growing number of PWR units that are switching from H-AVT to amine treatment for better protection of carbon steel compounds against FAC and similarly to decrease the iron concentration in feedwater that are deposited in the SG with many adverse effects.

A few PWR units operated with morpholine treatment at early times (e.g. Beaver Valley in the USA) but most of the French PWRs switched at early times to morpholine in 1984 for FAC mitigation, at least in all the units with copper alloys that had to operate at a pH₂₅ <9.3.

EPRI decided not to recommend morpholine and to compare various amines in the 1990's with preference for ethanolamine (ETA) in the 1990's. Other amines have also been studied and used in a few cases, but starting in the 2000's and continuing in the 2010's, morpholine and ETA have been the most widely used amines in PWRs and also in some VVERs where the plant results showed much better performances with amine treatment as compared to ammonia treatment.

Mainly in the USA, other amines are used in some cases, either alone or in a mixing of amines; including 3-Methoxypropylene (MPA) and Dimethylamine (DMA). Since these amines are not widely used and not considered here of having advantages over morpholine or ethanolamine, they are not discussed here.

Only the two main amines are compared between themselves and to ammonia, helping Utilities at selecting the best options for each situation and other organisms at understanding the advantages and inconveniences of each amine.

4 Deposits and Corrosion Products

4.1 Origin and consequences

As illustrated on Figure 4-1, the deposits in the SG, and on tubesheet may have either an external origin or in much larger quantities from internal origin.

The external origin can for example be the contaminants with a low solubility entering the secondary system from a condenser leak and precipitating in the SG due to the much lower solubility at high temperature as compared to low temperature. The salts containing calcium or magnesium have an extremely low solubility at high temperature. Thus, even without the concentration process described in section 3.4, they will easily precipitate and with the Hide out process, a very large fraction of these compounds is deposited in the SG.

On the opposite to soluble compounds like sodium, which will be mainly concentrated in low flow areas (crevices or sludge), calcium and magnesium may easily precipitate on SG tube surface, where the temperature is the highest and consequently the solubility the lowest. The elements of low solubility that are introduced into the secondary system through makeup water are of a marginal contribution.

The main origin of deposits and corrosion products is internal. The main contribution is coming from the generalised corrosion of carbon steel (CS) components, according to FAC described in section 2.4 or during lay-up. Then some other solid elements may be brought by various sources, such as resin fines during regeneration, metallic particles during construction, foreign objects during maintenance activities.

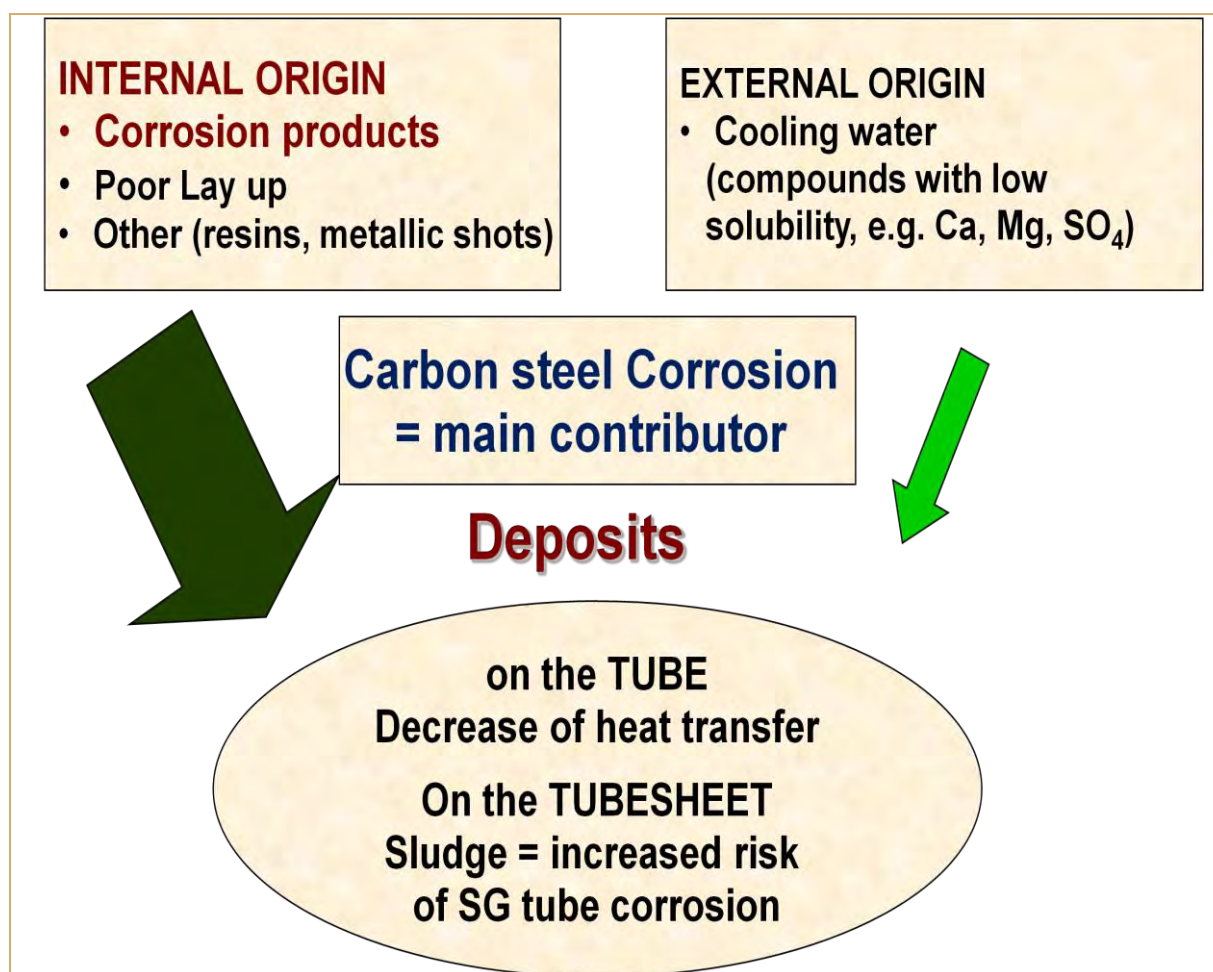


Figure 4-1: Origin of deposits in SG.

The most important contribution and point to optimise is CS corrosion that is largely covered in this report, in terms of FAC mitigation, materials selection and secondary side treatment optimisation. The iron oxides generated by FAC are entering into the SG where they have three possibilities:

- 1) They may deposit on the surface of the SG tubes, this is the fouling phenomenon that will decrease the heat transfer and potentially create a pressure drop and, for more severe cases, induce a power reduction;
- 2) They may deposit by gravity on the top of the tubesheet, forming the sludge pile, which may be more or less easily eliminated by sludge lancing during shutdown;
- 3) They may be eliminated by the SGBD, but usually with a limited efficiency.

The many consequences of deposits and corrosion products are schematised on Figure 4-2.

In the above case 1), the deposit on SG tube surface will deteriorate the heat transfer, which in turn, if too important, will induce a pressure drop and, for the most severe cases that cannot be compensated any more by valves opening, a power reduction. However, normally a remedial action to eliminate these deposits and restore the heat transfer before reaching such an extreme consequence, is implemented with one of the remedies described in section 4.2.

In the above case 2), the corrosion products will accumulate on the top of the tubesheet to form the sludge pile. The main consequence is the build-up of a location where the impurities may concentrate and induce corrosion as schematized on Figure 4-3. As explained in section 3.4.1, in the bottom of the sludge pile, water cannot access and this is a steam blanketed zone with hard deposits. At the top of the sludge pile, in the zone with enough water circulation, the concentration process is not an issue. In the intermediate zone below the top of the sludge pile, the water can access but not easily circulate and this is in this alternate wet and dry zone that the concentration of soluble impurities such as sodium will be the highest and may induce corrosion. The remedy described in section 4.2 usually consists in sludge lancing.

A key consequence is the recent observation of denting phenomenon at the top of the tubesheet and SG tube cracking detailed in section 2.3.3.2.

In the above case 3), the corrosion products that are as suspended solids will be partially eliminated by the SG blowdown. The efficiency of elimination by the SGBD depends on the location of the pipe, on the flow rate of SGBD and on the competition between the elimination and deposition either on the tube surface or in the sludge pile. The main remedy to increase the efficiency of the SGBD is the addition of dispersant described in section 4.2.4.

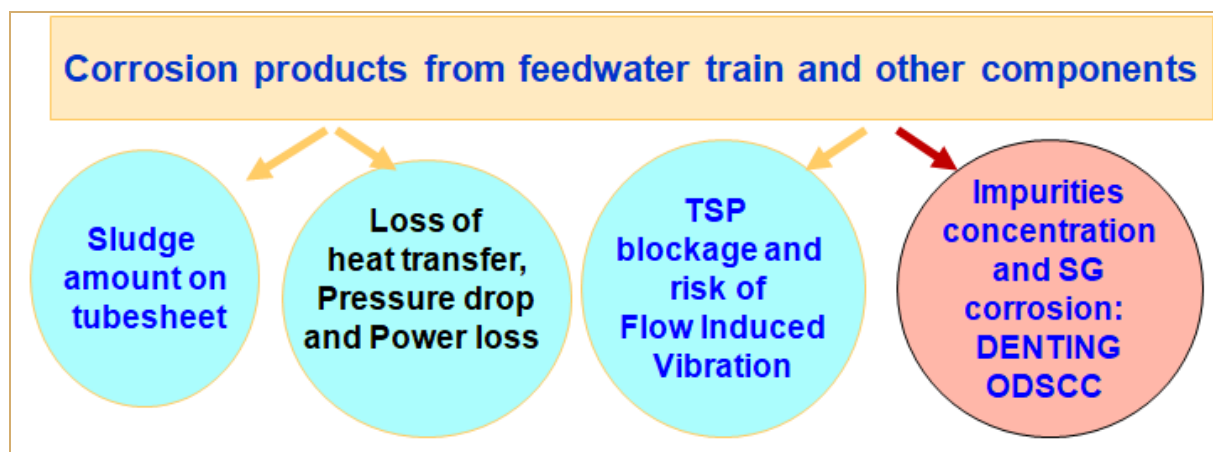


Figure 4-2: Main consequences of corrosion products and deposits.

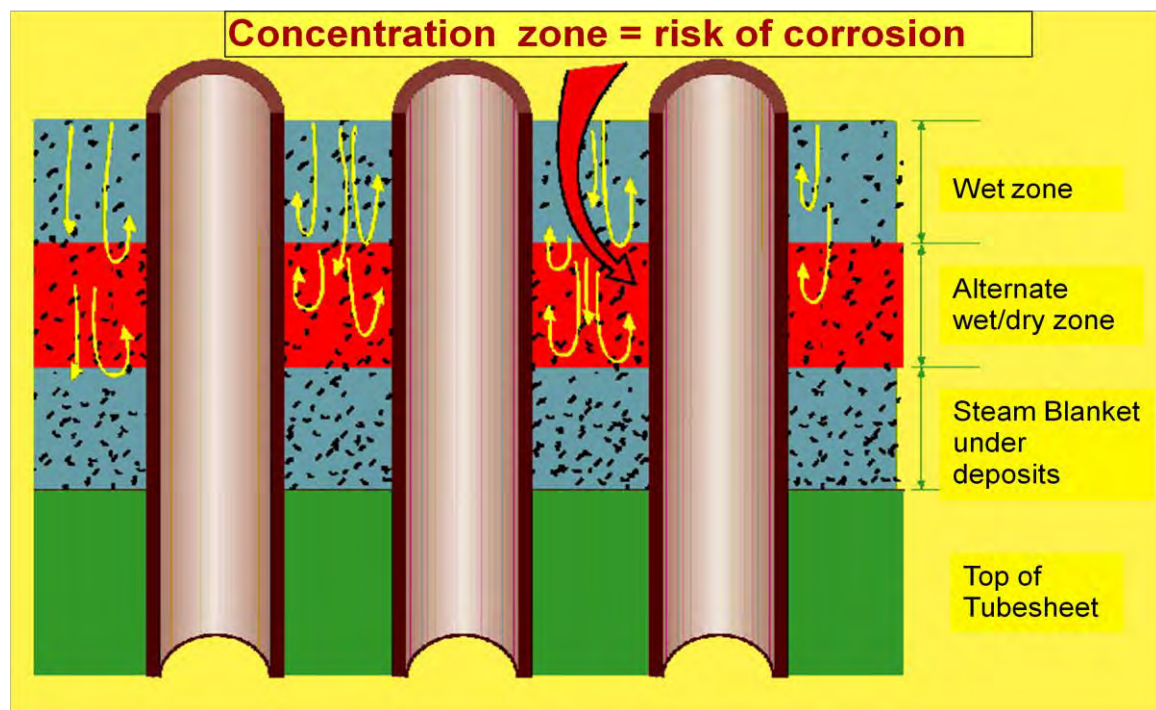


Figure 4-3: Concentration process and corrosion risk on sludge pile.

Then, another case, a mixing of case 1) and 2), has been encountered in several French units operating at a FW $\text{pH}_{25^\circ\text{C}}$ of 9.2 due to the presence of copper alloys in the condenser. These condensers were kept in copper to avoid proliferation of amoebae and legionella. Although the pH was maintained with amine treatment, it was not sufficient to prevent the transport of significant amount of iron oxides into the SG that deposited in local areas of broached TSP locations. This resulted in blockages of the broach holes by corrosion product deposits, as illustrated Figure 4-4.

Once the TSP is blocked, in some specific locations of holes without any tube, the high flow velocity induced Flow Induced Vibration (FIV) and circumferential cracking of the tube.

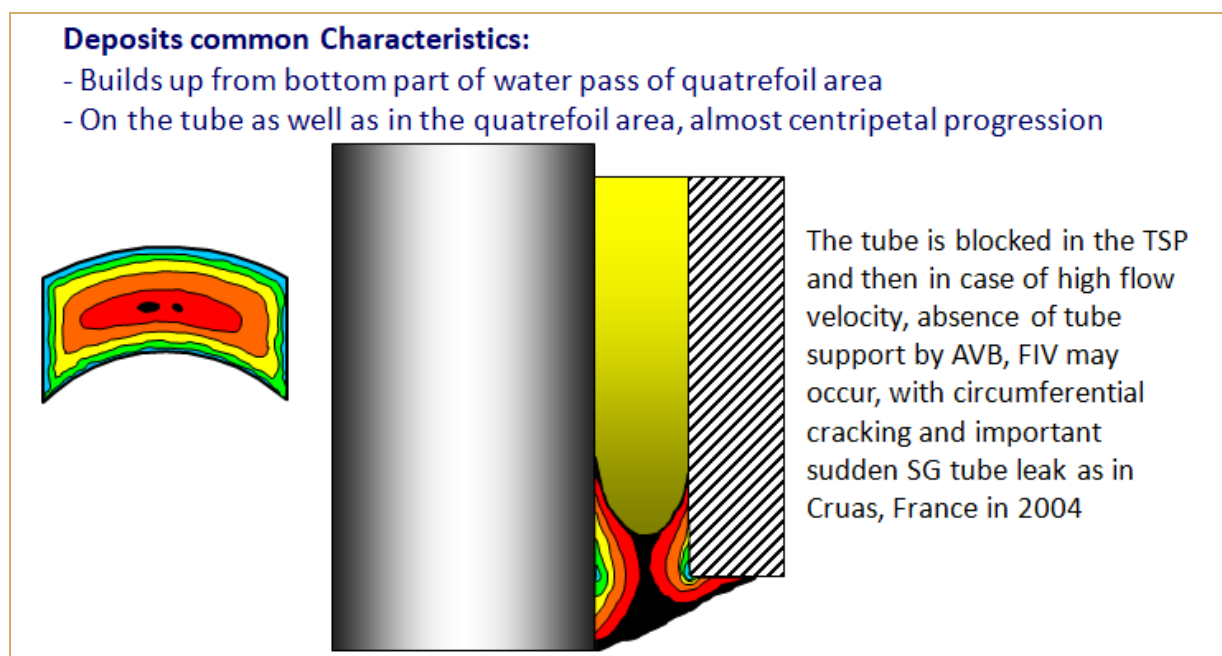


Figure 4-4: Deposits and Broached TSP Blockage in the French unit of Cruas.

4.2 Mitigation

There are preventive and curative ways of mitigating the deposits of insoluble elements, mainly made of iron oxides. The preventive ways are particularly acting on decreasing the amount of corrosion products that are in situ generated by selecting an appropriate secondary water treatment or in some plants by increasing the blowdown efficiency by addition of dispersants.

Of course, curative ways are applied either regularly if their burden and cost are acceptable, like the sludge lancing for eliminating the sludge pile, or occasionally if preventive remedies are insufficient. This is usually the chemical cleaning, with various types of process using chemicals in more or less amounts.

4.2.1 Materials pH, Treatment

The selection of the best treatment including the key objective of mitigating FAC, and consequently the amount of corrosion products transported to the SG has been largely described in section 3.2.

The first and easiest point is to select materials that are not sensitive to FAC, i.e. either stainless steel or at least low alloy steel, besides the condenser tubing material that may be titanium. Such a material selection applies to new NPP or older ones when revamped to eliminate carbon steel or more frequently copper alloys. It is cheaper and easier to replace copper alloys by other materials, respectively low alloy or stainless steel in heaters and MSR and by titanium or stainless steel in condenser. The absence of carbon steel applies mainly to new PRW units.

The absence of copper alloys is very important to allow the selection of a sufficiently high pH that minimises FAC rate of CS, as fully explained in section 3.2. In this section 3.2., it has been shown that either H-AVT treatment with ammonia at a $\text{pH}_{25^\circ\text{C}} > 9.8$ or amine treatment are able to mitigate FAC. Other remedies for FAC, such an oxygen addition, are also listed in section 2.4.

Several PWR or VVER units modified their secondary side treatment either to H-AVT or to amine treatment in years 1990's and 2000's.

As an example, the Swiss PWR of Beznau moved to H-AVT ($\text{pH}_{25^\circ\text{C}}$ 9.8-9.9) in 1993 and 1999 together with copper elimination. This allowed the feedwater Fe concentration to decrease from 7 to $2\mu\text{g/kg}$ (ppb) as shown on Table 4-1. With such an improvement, the Utility considered that there would no more the need for Chemical cleaning for the remaining life of plant.

Table 4-1: Inventory of magnetite in SG of Beznau after SGR [Mailand et al., 2012].

Unit (period)	Fe_3O_4 Ingress	Fe_3O_4 Removed	Fe_3O_4 Accumulated	Density g/m^2	Thickness μm
(1993- 2012)	510 kg/SG	180 kg/SG	330 kg/SG	100	20
(1998- 2012)	286 kg/SG	80 kg/SG	206 kg/SG	62	12

For PWR units with Condensate Polishing plants (CPP), H-AVT is not very suitable since it requires a high molar concentration of ammonia that implies a too frequent regeneration of IER. This is why several PWR units with CPP switched from ammonia treatment, that was not applied at a sufficient $\text{pH}_{25^\circ\text{C}}$, to amine treatment.

An example is shown in Table 4-2 for the Korean PWR unit of Ulchin-2 which switched from ammonia to ETA treatment, with a decrease in corrosion products of 55 to 90% in the whole steam-water system.

5 Present Issues of secondary system

This chapter is listing the present issues that are, in most cases, different from the old ones, particularly due to the evolution in SG tubing materials.

The key points are minimising the amount of corrosion products and deciding on the best purification systems allowing to get a sufficient pH and consequently a low amount of corrosion products.

Then, a specific section discusses the controversial influence of lead in encountered degradation on SG tubes and on future remedies.

Finally, a section is summarising the key facts for each type of SG tubes in order to ensure a long plant life operation.

5.1 Corrosion products

The issues associated with corrosion products have been described in details in sections 2.2, 3.2 and 4.

The reason to list this technical point here is to highlight the fact that the presence of corrosion products has not been handled properly at the time when the focus was on the presence of soluble impurities that were considered as the direct cause of corrosion on the secondary side of SG tubes, particularly with the highly sensitive Alloy 600 MA.

The Figure 5-1 below allows to understand that this issue is becoming of growing importance for most of the PWR and VVER units with Alloys 600TT, 690TT, 800NG, or 18-10-Ti since SG with Alloy 600MA are disappearing with their replacement or the definitive shutdown (a few US cases) of the units with such SG tubes.

As already explained in section 3.2, corrosion products are coming from corrosion of carbon steel, and this is even more crucial when the pH is too low due to the presence of copper alloys or of condensate polishing plants to avoid a frequent regeneration (see section 5.2) or of a low oxygen for trying to mitigate SCC of SG tubes.

The consequences of these corrosion products have been listed in section 4.1:

- (1) deterioration of the heat transfer,
- (2) pressure drop and, in the most severe cases,
- (3) power reduction,
- (4) build up of a sludge pile where the impurities may concentrate and induce corrosion,
- (5) denting phenomenon at the top of the tubesheet.

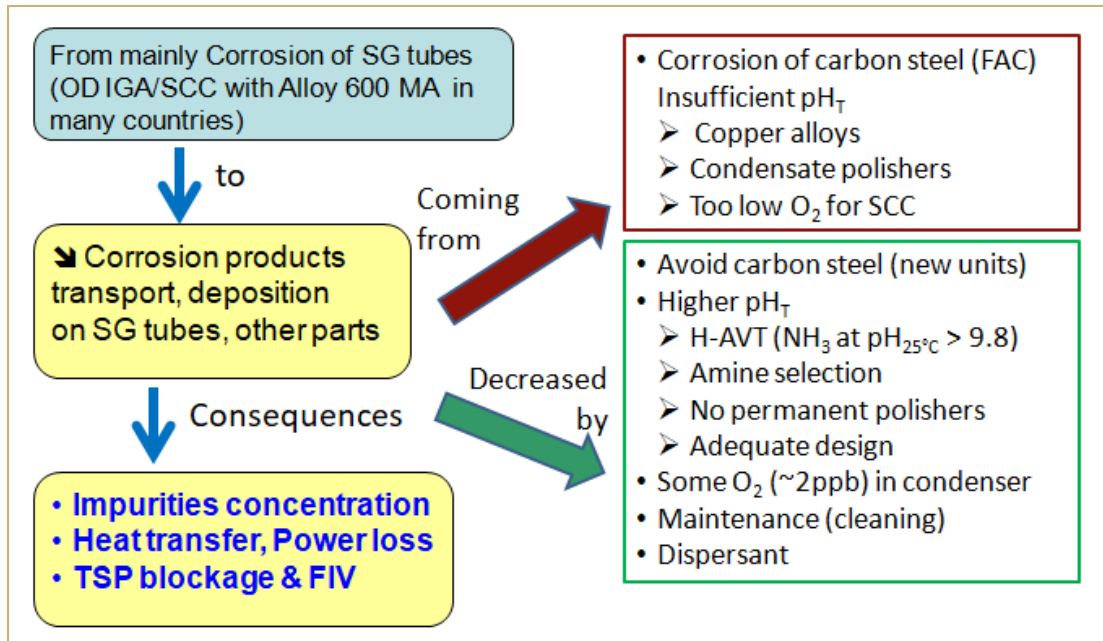


Figure 5-1: Corrosion products origin and remedies to decrease them.

5.2 Wastes, Operating costs – CPP and SGBD options

A Condensate Polishing Plant (CPP) is installed for purifying the water coming from the condenser, which, after the FW train, will be the FW for the SG. The CPP is not always necessary and the decision to install it or not and to use it or not depends on various criteria, some of them being based on old situations when the condensers were not tight enough to allow a sufficient purity in the SG water.

Then, even with tighter condensers, some Utilities decided to have CPP to be sure to comply with specifications at SGBD, particularly when relying on EPRI guidelines that have been settled at very restrictive values due to the presence of sensitive Alloy 600MA (see section 2.2).

It consists in a sequence of Ion Exchange Resin (IER) beds and/or filters with powdered resins, to eliminate corrosion products and ions present in condensate water i.e. ~55% to 2/3 of the FW.

Generally, there is a cation bed for conditioning reagent elimination followed by a mixed bed (polishing) to better eliminate, in a neutral environment, the final traces of impurities coming from the condenser in case of leak. Most of CPP include traditional regenerable resins, called deep beds, but there are, in the USA, some stations with powdered resins (Powdex) to insure an efficient filtration.

The CPP is followed by filters to avoid release of resins fines into the FW and the SG where they would be highly detrimental for SG tubing corrosion due to the presence of sulphur compounds.

There are several advantages and inconveniencies in using CPP.

The benefits of having a CPP are:

- To continue operation with a leaking condenser when this is a river water of low or intermediate condenser leak rate or when this is a seawater with low leak rate below the threshold to be localisable (< 0.05 l/h).
- To allow a better decision on the best moment for a shutdown with the dispatching in case of leak.
- To avoid the large pollution from an important condenser leak to reach the SG; however, this may also be avoided or minimized without a CPP with quick action.
- To partially or totally replace the SGBD purification treatment.
- To allow a quicker start up after shutdown.

The numerous disadvantages of CPP are:

- High investment costs.
- High operating costs for chemicals, resins and the time spent for regenerations and monitoring.
- Important liquid effluents into the environment, which is one of the most important concerns for the future, sustainable development.
- Does not manage all condenser leaks (e.g. large seawater leaks).
- The highly detrimental consequence of CPP in permanent operation is the impact on the difficulty to apply a sufficiently high molar concentration of alkaline reagent and thus the impossibility to have a sufficiently high pH_r, that is necessary to minimise FAC of carbon steel and transport of corrosion products to the SG. This limitation for the reagent concentration comes from the associated frequency of regeneration of Ion Exchange Resins (IER) of the CPP.

As confirmed by the EPRI survey shown on Figure 5-2, CPP (deep beds) is detrimental for corrosion product. Due to pH limitation, the generation of corrosion products downstream the CPP is not compensated by elimination of oxides generated upstream the CPP.

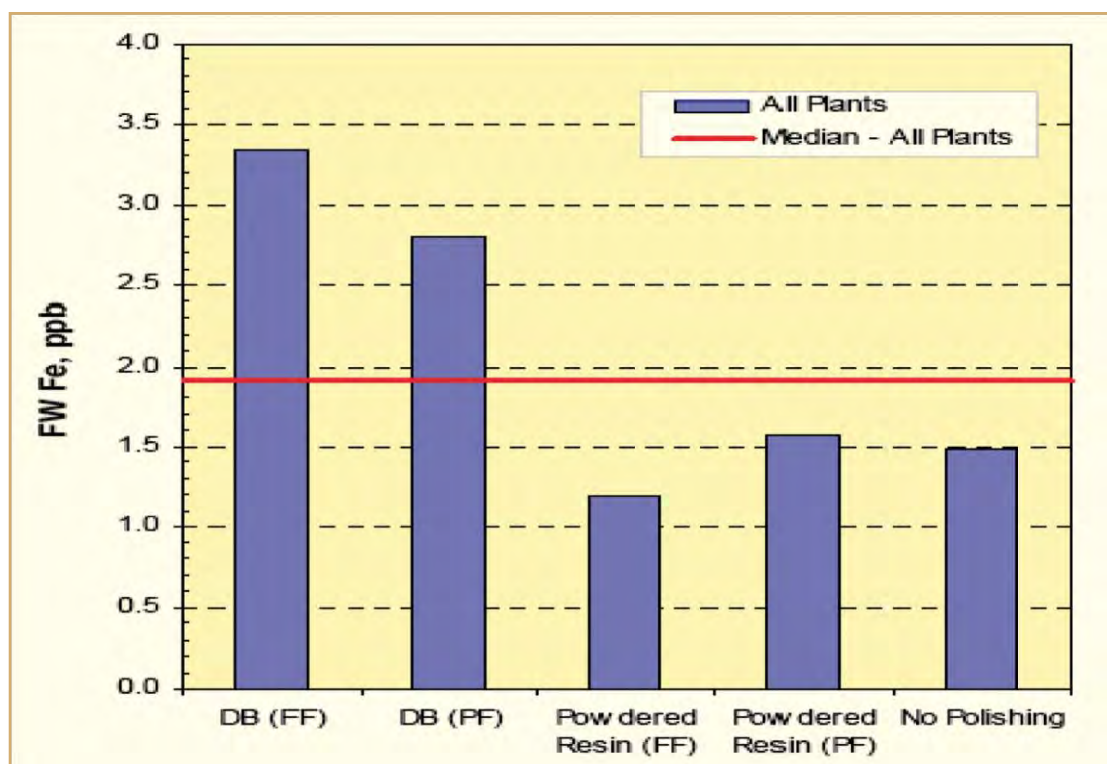


Figure 5-2: Detrimental impact of CPP on Fe in FW [Choi et al., 2008].

In the same EPRI survey shown on Figure 5-3, CPP in US plants does not allow significantly lower values of soluble impurities (Na, Cl, SO₄) at SGBD, demonstrating that the main target of CPP is not even achieved. Anyhow, even without CPP, the values of ions at SGBD are sufficiently low for the resistant materials such as Alloy 800 NG and Alloy 690TT.

Most part of the time, without leak, this is even more a pollution source than a purification mean; purely alkaline impurities, such as sodium throw at CPP outlet are extremely detrimental for SG tube with Alloy 600 MA (much more than a condenser leak) as noticed in Spain, where SG were highly corroded after operation with ~10-20 µg/kg of alkaline sodium at SGBD, coming from CPP pollution.

Finally, the risk of pollution by resin fines and regeneration reagents can cause significant SG tubing corrosion, due to the sensitivity of all SG tubing materials to sulphur compounds (resin fines) and strongly acidic or alkaline environments (reagents).

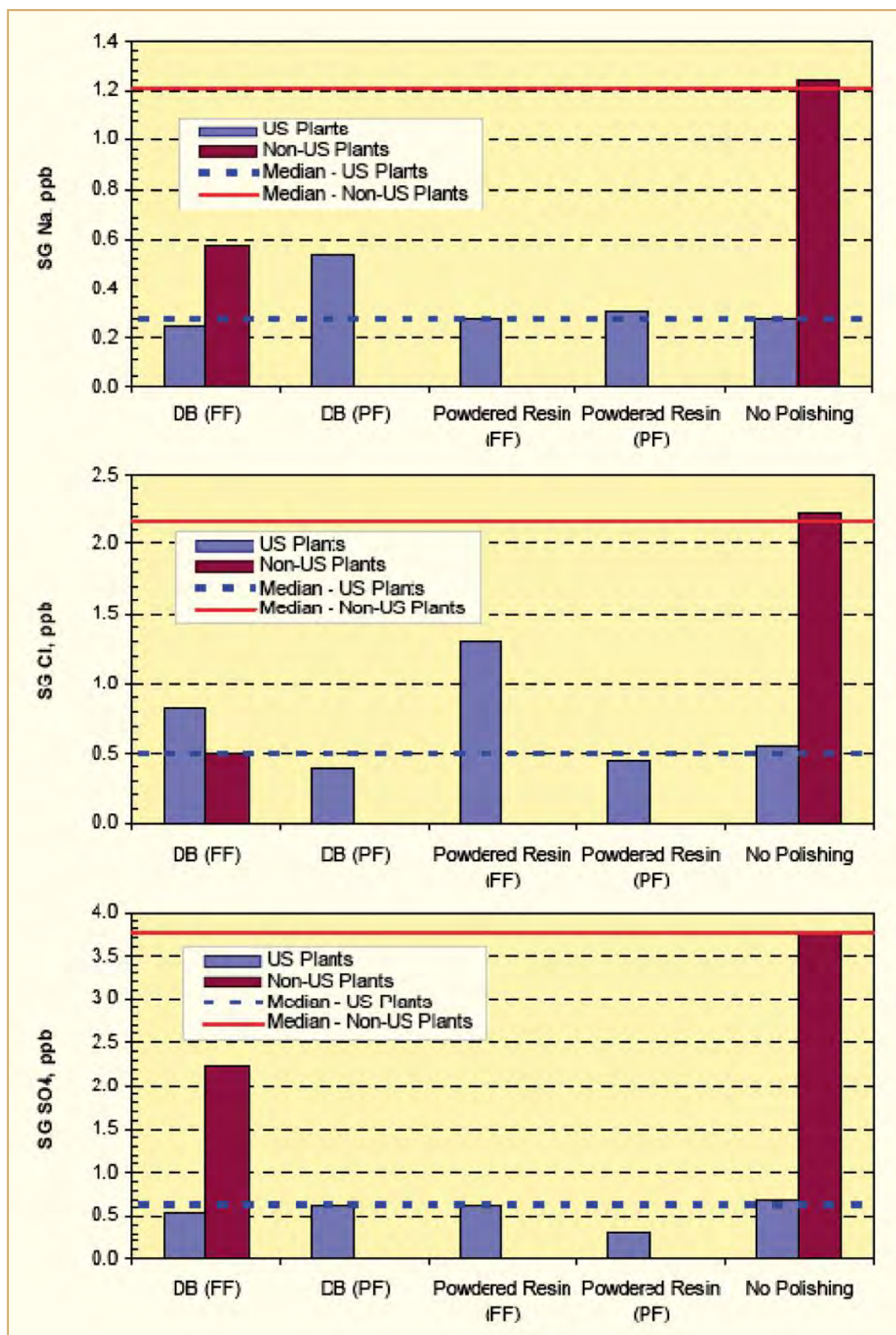


Figure 5-3: Influence of CPP on sodium, chloride and sulphate at SGBD [Choi et al., 2008].

6 Conclusion

In conclusion of this survey of chemistry and corrosion in PWR and VVER units, according to the existing materials, it is extremely important to define the corresponding chemistry treatment and specifications. Unfortunately, in many cases, these specifications do not enough take into account the evolution of materials and focus on old issues that are not any more the most important ones.

This is why this conclusion gives the key information from this document, with different conclusions for different SG tubing materials.

In a first part, the evolution of treatments explains that the initial phosphate treatment that was applied to buffer impurities from condenser leak, was not any more appropriate with units of higher heat flux and temperatures where the reagent was concentrating and inducing either alkaline or acidic corrosion in local areas. In addition, with the evolution of condenser design with titanium tubing, the number of condenser leaks has drastically decreased. German units, which also kept phosphate treatment for a longer time, also switched to AVT treatment due to wastage of SG tubes.

An early important modification in materials selection has been the elimination of copper alloys from the entire secondary system and their replacement for the condenser either by titanium tubes or stainless steel. This modification allowed to select a better secondary side AVT with a higher $\text{pH}_{25^\circ\text{C}}$ chemistry, either with ammonia at high pH (9.8-10) or with amine treatment, mainly morpholine or ethanolamine (ETA).

Plants still having copper alloys with an upper limit of pH of 9.2 should not have remained with ammonia treatment in presence of carbon steel. French units switched early enough to amine treatment (in 1984) with morpholine to mitigate Flow Accelerated Corrosion (FAC) of carbon steel. Several American plants moved later on to amine treatment with ethanolamine in the 1990's. German plants and some other plants moved to H-AVT (ammonia at $\text{pH} > 9.8$).

However, some Japanese units remained at low pH with ammonia, due to the presence of Condensate Polishing Plants (CPP) that are not compatible with a high reagent concentration, which would induce a high Ion Exchange Resin (IER) regeneration frequency. A tragic consequence of this decision, was a significant FAC rate, associated with insufficient monitoring of CS thickness decrease, was a burst of a pipe that caused several deaths and injuries within workers. Then after the Japanese units, like many other units in various countries, selected either amine treatment, mostly ETA or H-AVT.

VVER units, also with CPP, moved slowly and later on to amine treatment.

Nowadays, there are consequently two types of secondary side treatments that are suitable, H-AVT at a $\text{pH} > 9.8$ or amine treatment. In presence of copper alloys, amine treatment will be applied with a pH limited to 9.2 and in the absence of copper alloys, the pH and amine concentration will be increased as necessary to minimise FAC of carbon steel. This pH value and amine concentration will depend on many factors such as wastes releases restriction, purification systems, extent of CS components:

- H-HVT, which has for main advantages the simplicity of implementation and control, the absence of decomposition products but for main inconveniences, the need to operate at a high molar concentration of ammonia, hardly compatible with CPP and requiring a high frequency of IER regeneration.
- Amine treatment with ETA, which has for main advantages a good protection for FAC, a large feedback, a low molar concentration, some compatibility with CPP, but for main inconveniences, a limited possibility of operating with IER saturated by the reagent and a questionable behaviour on IER.
- Amine treatment with morpholine, which has for main advantages, also a good protection for FAC, the possibility to operate IER at SGBD saturated with the reagent, thus with low frequency of IER regeneration or even the total absence of regeneration, avoiding any risk of contamination, but for main inconveniences, a limited possibility of operating with IER saturated by the reagent, production of organic acids, and a higher cost of reagent.

In addition to the alkaline reagent, hydrazine, a reducing agent is also injected in the secondary system. Based on the plant experience feedback and in agreement with most of existing documents, guidelines and specifications, the proposed limit value of hydrazine concentration is $\geq 20 \mu\text{g/kg}$ and $\geq 8 \times [\text{O}_2]$ in the condensate water. More generally, a target value of $50 \mu\text{g/kg}$ seems optimal if it is acceptable with the upper pH limit.

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List of Abbreviations

600MA	Alloy 600 Mill Annealed
600TT	Alloy 600 Thermally Treated
690TT	Alloy 690 Thermally Treated
800NG	Alloy 800 Nuclear Grade
AISI	American Iron and Steel Institute
ANTI	ANT International
ASCA	Advanced Scale Conditioning Agent
ASME	The American Society of Mechanical Engineers
AVB	Anti Vibration Bar
AVT	All Volatile Treatment
CE	Combustion Engineering
CGR	Crack Growth Rate
CPP	Condensate Polishing Plant
CS	Carbon Steel
DART	Deposit Accumulation Reduction Treatment
DEHA	Diethylhydroxylamine
DMA	Dimethylamine
DMT	Deposit Minimization Treatment
DO	Dissolved Oxygen
ECP	Electrochemical Potential
EDA	Ethylene-diamine
EdF	Electricité de France
EDTA	Ethane-1,2-diylldinitrilo) tetra acetic acid
EPR	European Pressurized Water Reactor
EPRI	Electric Power Research Institute, USA
ETA	Ethanolamine
FAC	Flow Accelerated Corrosion
FFA	Film Forming Amines
FIV	Flow Induced Vibration
FW	Feed Water
H-AVT	AVT with ammonia at high pH

Unit conversion

TEMPERATURE		
$^{\circ}\text{C} + 273.15 = \text{K}$	$^{\circ}\text{C} \times 1.8 + 32 = ^{\circ}\text{F}$	
T(K)	T($^{\circ}\text{C}$)	T($^{\circ}\text{F}$)
273	0	32
289	16	61
298	25	77
373	100	212
473	200	392
573	300	572
633	360	680
673	400	752
773	500	932
783	510	950
793	520	968
823	550	1022
833	560	1040
873	600	1112
878	605	1121
893	620	1148
923	650	1202
973	700	1292
1023	750	1382
1053	780	1436
1073	800	1472
1136	863	1585
1143	870	1598
1173	900	1652
1273	1000	1832
1343	1070	1958
1478	1204	2200

Radioactivity	
1 Sv	= 100 Rem
1 Ci	= 3.7×10^{10} Bq = 37 GBq
1 Bq	= 1 s^{-1}

MASS	
kg	lbs
0.454	1
1	2.20

DISTANCE	
x (μm)	x (mils)
0.6	0.02
1	0.04
5	0.20
10	0.39
20	0.79
25	0.98
25.4	1.00
100	3.94

PRESSURE		
bar	MPa	psi
1	0.1	14
10	1	142
70	7	995
70.4	7.04	1000
100	10	1421
130	13	1847
155	15.5	2203
704	70.4	10000
1000	100	14211

STRESS INTENSITY FACTOR	
MPa $\sqrt{\text{m}}$	ksi $\sqrt{\text{inch}}$
0.91	1
1	1.10