



# LCC8 ANNUAL REPORT

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

# Key Emerging Issues and Recent Progress Related to Plant Chemistry/Corrosion

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# 1 Introduction (Francis Nordmann and Dewey Rochester)

The NPC 2012 conference took place in Paris 24-27 September 2012. It was the 18<sup>th</sup> conference in the series of “Nuclear Power Plant Chemistry” that started in Bournemouth in 1977 and is now taking place every other year. The previous one, NPC 2010, was in Quebec, and the conclusion and key facts were presented at LCC6 seminar while the next one will be in Sapporo in 2014.

Over 350 participants from 28 countries attended this conference, the highest participation in the history of these conferences, despite the fact that there are some uncertainties after Fukushima event in March 2011 and the decision of some countries to shut down their Nuclear Power Plants (NPP) in the medium or long term. However, on the opposite, there are new programs of building new NPP in some other countries (mainly China, UK, Eastern Europe, India, etc.).

There were 267 attendees from Europe (mainly France of course, but also with a significant participation from various other European countries. Then, there was respectively 46 attendees from Asia, 37 from America and 4 from Africa.

There have been 15 sessions where 62 papers have been presented and 3 sessions with 140 posters.

D. Minière, Electricité de France (EDF) Executive Vice President, Head of Nuclear Power Plant Operation (58 reactors) made the opening speech explaining that there are > 400 reactors in the world. France, thanks to its large fleet, is producing 95% of its electricity with almost no CO<sub>2</sub> releases (nuclear, renewable including hydraulic) and also the cheapest in Western Europe. Then he focused on the need to implement the lessons learned from Fukushima accident in order to produce Nuclear Energy without uncontrolled releases of radioactivity if we want a public acceptance of this energy. It is our community responsibility to avoid such accidents and take into account the increasing impact of tropical storms, e.g. the first one in France occurred in 1999. Consequently, the Nuclear Power Plant (NPP) robustness must be increased with an adequate preventive design, including resistance to flooding particularly in countries where such storms may occur in large number and intensity.

We also need to face the unforeseeable thanks to robust organizations and particularly being able to restore water and electricity within the first 24 hours. International Atomic Energy Agency (IAEA) capability should be supported and World Association of Nuclear Operators (WANO) Peer Reviews within Operators must be more accurate with a large safety focus. Regulators should also establish exchanges in nuclear safety.

Together with design modification, chemistry is one aspect contributing to enhance safety and life extension with an increased reliability, a decrease of Operational Radiation exposure (ORE) a good radioprotection.

One of the key aspects is the Steam Generator (SG) cleanliness thanks to an adequate secondary system treatment.

The third Steam Generator Secondary Side Management Conference, EPRI SGMP 2012, was held in Atlanta, GA USA, on September 11-13, 2012. The first two conferences were respectively held in February 2003 in Savannah, Georgia USA and in March 2010 in San Antonio, Texas USA

There were 82 participants at the conference with 60 from the U.S and 22 internationals and 6 Exhibitors. The non U.S. participants were mostly from France (10) and Canada (5). Finland, Germany, Slovenia, Korea and Sweden were also represented. The Exhibitors were Rolls Royce, GE Power and Water Process Technologies, Puro-lite, Swan Analytical Instruments, AREVA and Westinghouse [Choi, 2012].

The goal of the conference was to facilitate exchange of information on:

- Current events and management issues
- New technology developments

The conference was divided into four sessions plus a keynote introductory presentation.

- I. Session 1 – Deposit Generation and Transport
- II. Session 2 – Deposit Control and Mitigation
- III. Session 3 – Deposit Consolidation and Removal
- IV. Session 4 – Short and Long Term Strategic Planning

## 2 Primary water chemistry (NPC 2012) (Francis Nordmann)

### 2.1 General information

The new VVER-TOI design provides improvement of safety, technical-economical aspects and operation, allowing for example the load follow. The main features are directed towards lower dose rates with:

- Suppression of oxidizing radiolytic products and activated corrosion products;
- Better  $\text{pH}_T$  stability also for decreasing Occupational Radiation Exposure (ORE);
- Direct addition of hydrogen instead of producing it through ammonia like in previous VVER designs, which also allows a better  $\text{pH}_T$  stability;
- Zinc addition depleted in zinc-64, only for VVER-TOI, considering that the presence of copper alloys in VVER already in operation is bringing some zinc [Susakin & Brykov, 2012].

The possibility of load follow has been taken into account. If ammonia has been given up for direct hydrogen addition, like in PWR design, the other key difference in chemistry will not change and the  $\text{pH}_T$  will still be obtained with potassium injection instead of lithium. Consequently, the total alkali (K + Na + Li) will still be monitored and is presented in various zones, as shown on Figure 2-1. The similar zones for VVER-1000 units are shown on Figure 2-2.

Surprisingly, only chloride remains as an anion control parameter (< 0.1 mg/kg) while sulphate is only a diagnosis parameter.

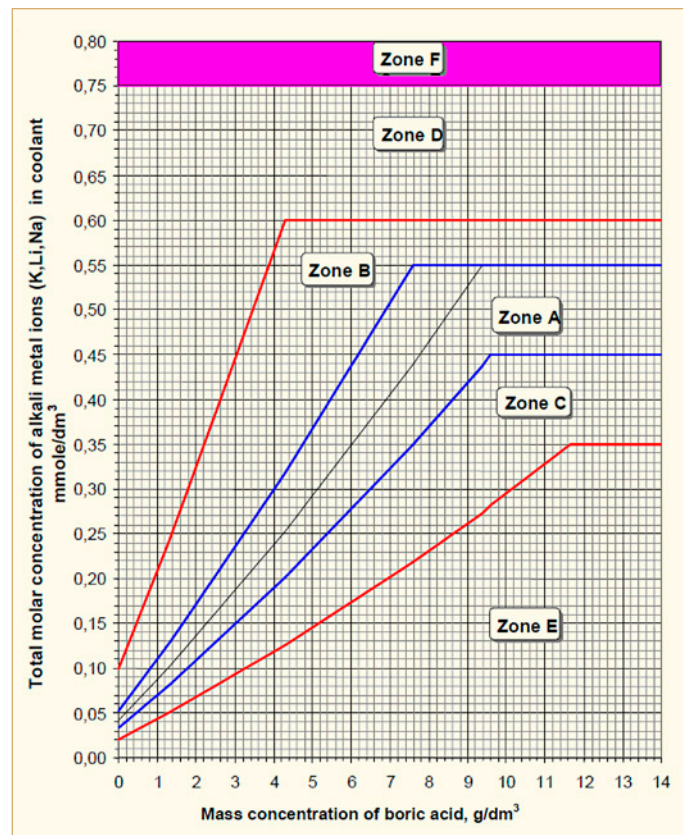


Figure 2-1: Total molar concentration of alkali (K + Li + Na) vs. Boron in VVER-TOI [Susakin & Brykov, 2012].



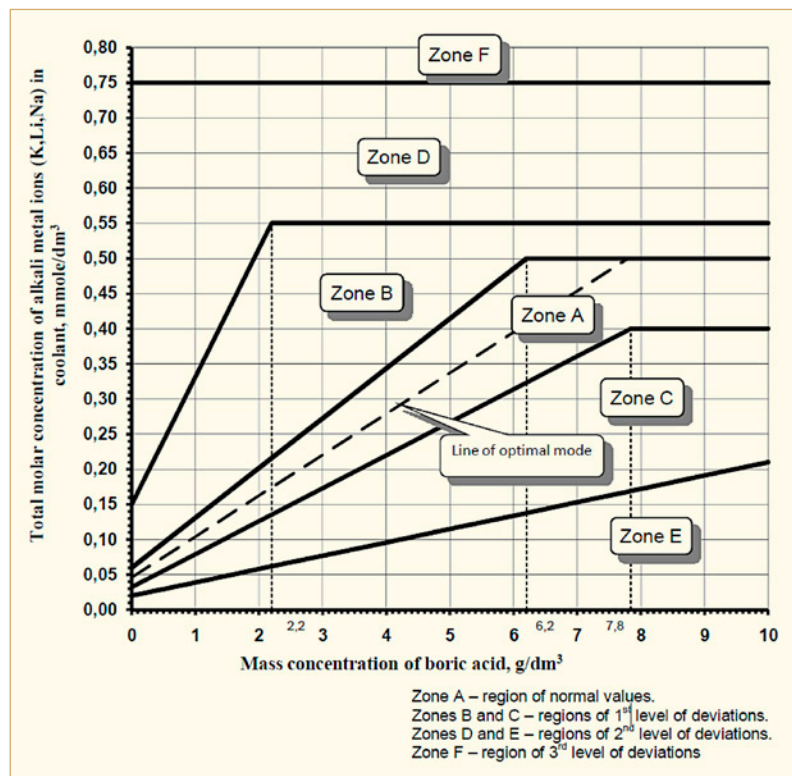


Figure 2-2: Total molar concentration of alkali (K + Li + Na) vs. Boron in VVER-1000 [Susakin & Brykov, 2012].

## 2.2 PWR, VVER radiochemistry and dose rates

EDF and CEA jointly performed an interesting comparison of dose rates within several PWR units of several countries (France, UK, Belgium and Spain), with EMMEC campaigns. The interesting aspect of such a comparison is to evaluate the respective influence of various factors such as the SG Alloy tubing, the tubing cobalt content, components with Inconel other than the SG presence of stellite, duration of maintenance activities, organization improvement, chemistry in operation.

From the results of Figure 2-3, it can be seen that Sizewell, which had an excellent HFT procedure is showing good results. It is interesting to compare the two Belgian units Doel 3 where the SG replacement (SGR) was done in 1993 with Alloy 800NG to Doel 4 where the SGR was done with Alloy 690 TT in 1996, both units having the same chemistry (pH 7.2 DH in the 25-35 cc range) but respectively 12 and 18 month fuel cycles. The higher dose rate in Doel 3 may be explained by more Cr-Ferrites in the passive layers in presence of Alloy 800 than in case of nickel base alloy (Alloys 600 or 690) as SG tubes, and the higher affinity of Co for incorporation in Cr-Ferrites than in Ni-ferrites. Trillo, in Spain, has Alloy 800 SG tubing. Co-60 and Co-58 being the main contributors to dose rates, the chemical elements to be limited or for which the release should be minimized are cobalt and nickel. The main potential reasons for higher <sup>60</sup>Co are SG tubing content, stellites and Inconel grids, although the results do not show lower values for SG having the lowest Co content. It is very difficult to draw any firm conclusion from these various parameters and Inconel grids may potentially be an important one. The main potential reason for lower <sup>58</sup>Co is the use of Alloy 800 tubing which contains less nickel than Alloy 690, however, it is obvious that this is not the main parameter for low dosimetry which is shown by Doel 4 with Alloy 690; there are a lot of beneficial parameters, individually necessary but not sufficient to improve contamination behaviour. No chemistry parameter seems to have an impact on dose index, which can be evidenced, and it may be concluded that their influence is lower than other factors. Finally, the organization of maintenance activities during outages is likely an important point that may reduce dosimetry, more than the spent time. For expelled a good preparation and association, of a Health Physician together with the working team is important [Guinard et al, 2012].

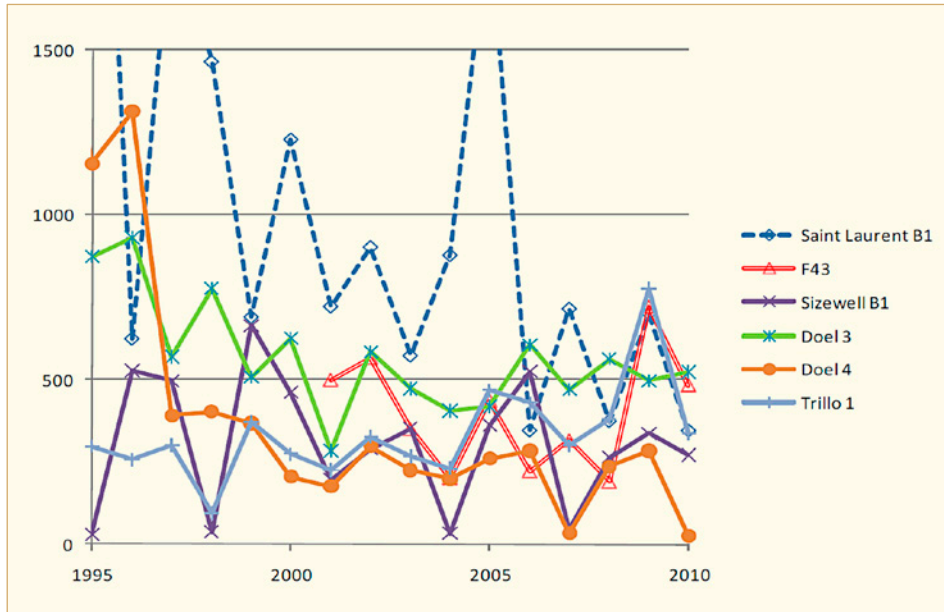


Figure 2-3: Comparison of collective dosimetry (Man. mSv) in various PWR units where F43 are French 1450Me sister units [Guinard et al, 2012].

In a poster paper, EDF described the overall project of source term reduction which includes: contamination prediction and optimization tools (corrosion and fission products, metal releases from SG tubes), zinc injection, improvement of operating procedures including shutdown and start-up, of purification media (filters, IER, other innovative media), of materials (stellites, silver, antimony, electro polishing, pre-oxidation + cleaning before start-up, SG tubes manufacturing).

EPRI presented a survey of chemistry practices in PWR units including the influence of zinc, depleted or not represented on Figure 2-4 [Perkins et al, 2012].

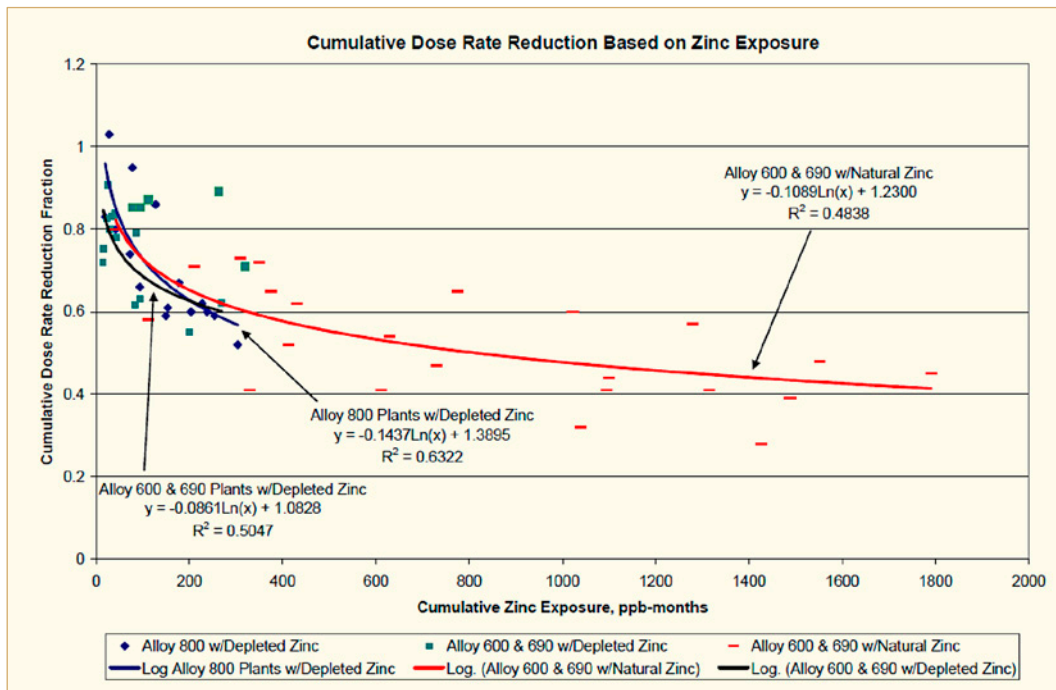


Figure 2-4: Influence of zinc addition on dose rates in PWR units [Perkins et al, 2012].

The radiochemical process for radiochemical specifications during shutdown at EDF is focusing on forced oxygenation by the injection of  $H_2O_2$  at  $80^\circ C$  for controlling the solubilisation of activated corrosion products, ensuring their purification at the highest rate and securing the absence of hydrogen in the RCS.

An American study on corrosion product releases from SG tubing has been performed on over 20 units (18 units with Alloy 690 after SGR) with the objective of studying the impact on corrosion products deposition on the fuel with the typical potential risks: (i) fuel cladding corrosion, (ii) boron precipitation and thus AOA, (iii) dose rates from activation. There are plants where the corrosion / release rate is decreasing sharply after being in service and those with a constant rate as illustrated by 3 different plants on Figure 2-5 [Marks et al, 2012].

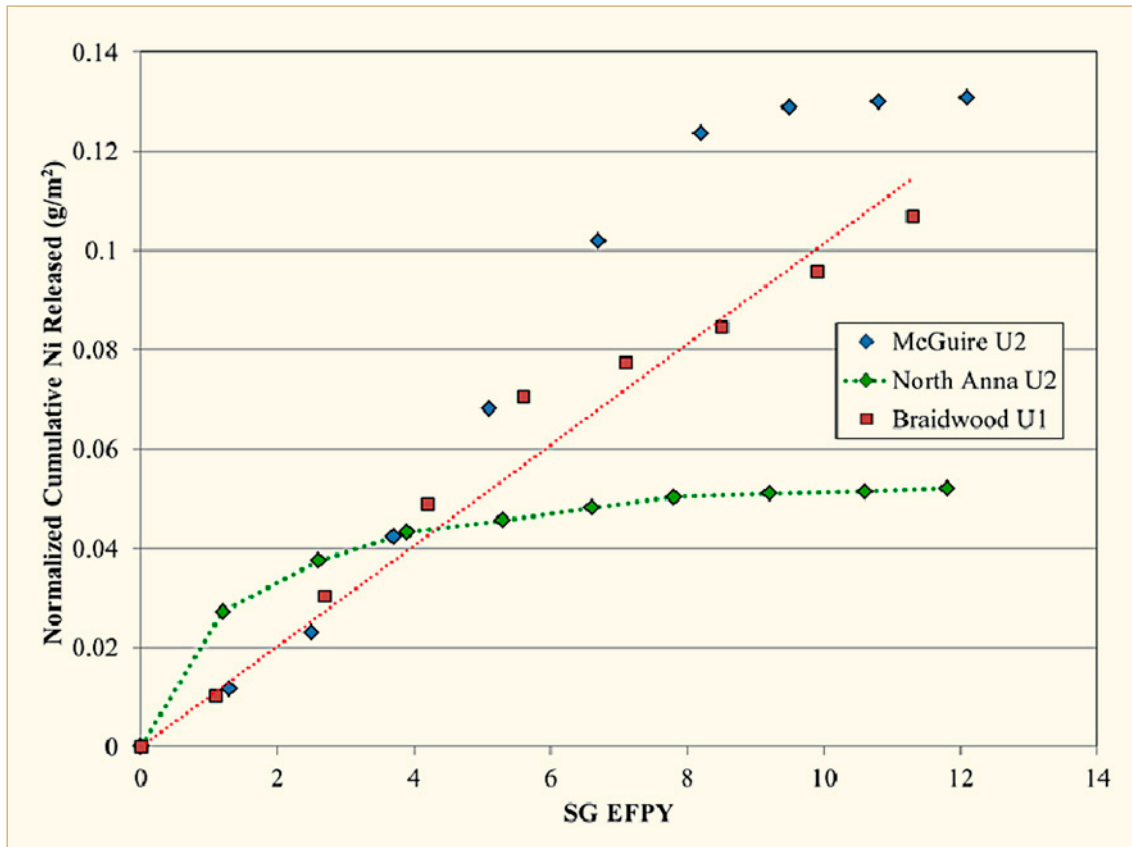


Figure 2-5: Comparison of cumulative nickel released (Normalized by SG area) vs.; EFPY for different trends with time [Marks et al, 2012].

It appears that the key parameter, among those observed (zinc addition, plant type, temperature, SG tubing) is the tubing manufacturer as observed in other studies (mainly in France) as shown on Figure 2-6. The second most important parameter is the temperature and no clear correlation has been made with  $pH_T$  of the RCS. Zinc addition has been looked at since it belongs to the chemistry factors, but as expected, it is playing a role in Co replacement on the deposits on the fuel but not apparently on the corrosion / release rate from SG tubing. If any influence of  $pH_T$  or other chemical parameter including zinc, it is hidden by the manufacturer much stronger effect. However, it seems that units that added zinc before SGR had the lowest long-term release rate. [Marks et al, 2012].

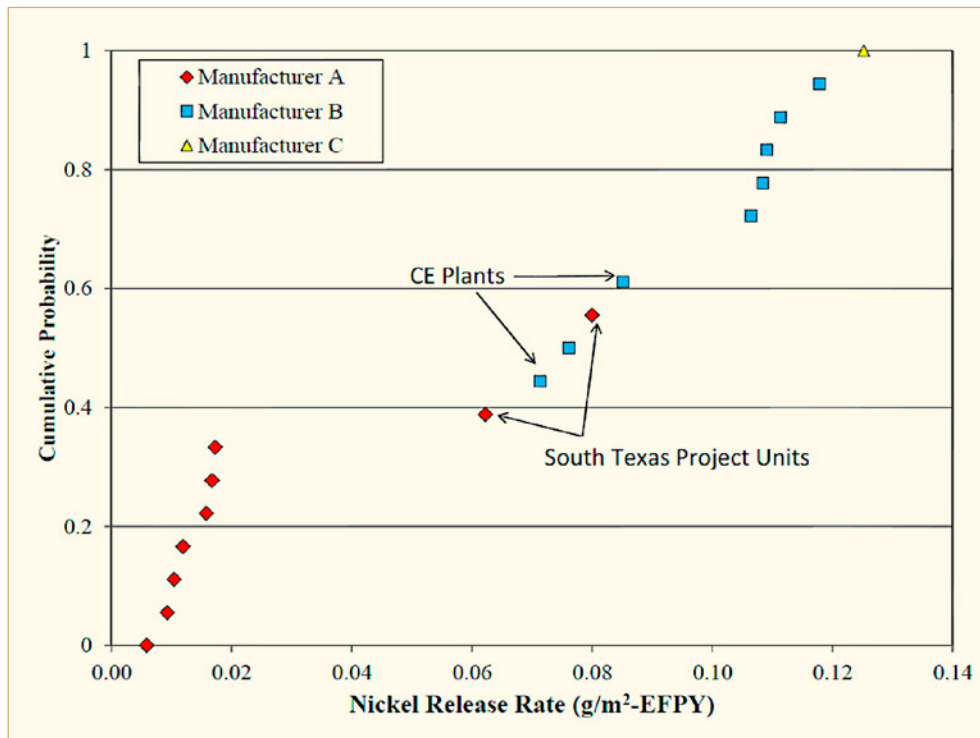


Figure 2-6: Cumulative probability distribution of long term Nickel release rates separated by tube manufacturer [Marks et al, 2012].

As shown on Figure 2-7, there has been a large reduction in shutdown releases after cycle 4 of Sizewell B1 (UK), which is believed to be due to the passivation of SG tubing, with a very low radiation field in SG, although the SG channel heads were not electro-polished and that zinc has not added in the plant [Barton et al, 2012].

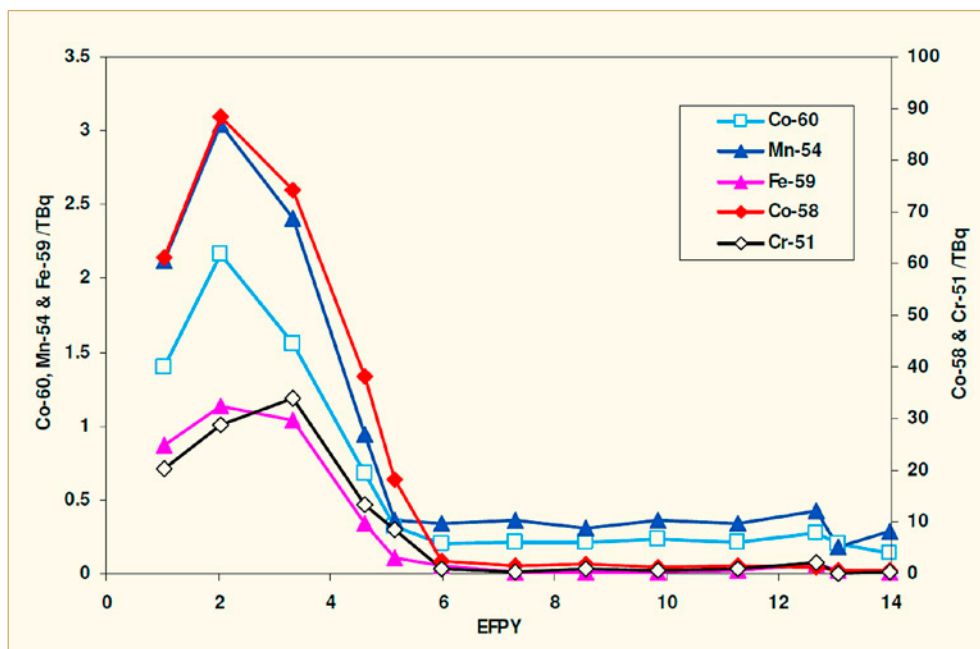


Figure 2-7. Integrated shutdown releases at Sizewell B, up to RO 11. RF07 corresponds to 8.55 EPY, FO28 to 513 EPY, FO43 to 13.05 EPY and RO11 to 13.97 EPY [Barton et al, 2012].

As shown on Figure 2-8, among three surface treatment of nickel base alloys for decreasing further release of nickel and thus dose rates, the pre-oxidation in humidified H<sub>2</sub> was more efficient than electro-polishing or pre-oxidation in high temperature water. The investigation was made by electrochemical methods [Kanzaki et al, 2012].

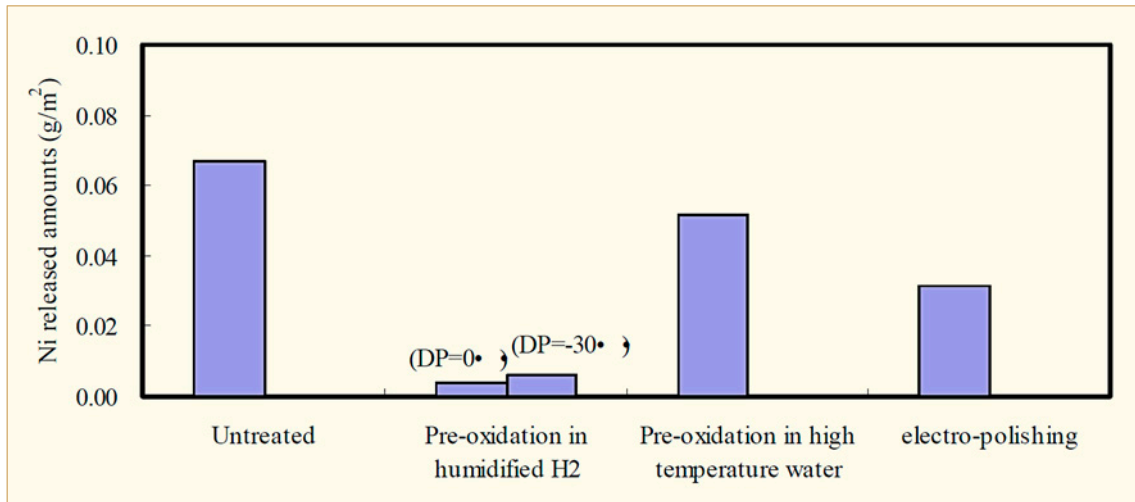


Figure 2-8: Results of nickel release test for various surface treatment [Kanzaki et al, 2012].

A British Laboratory, in cooperation with EPRI and the feedback from 2 PWR units (Sizewell B and Vandellos) developed a model to evaluate the behaviour of corrosion products (soluble Ni and Fe particulate form) and their activation. It is considered that when the alloys corrode, half is forming the inner chromite oxide layer and the other half may be either released to the coolant or form the outer particulate layer. The uptake of activity is occurring with passage from soluble radionuclide to inner oxide or precipitation on outer layer. Comparison of the model with plant data has been made and seems satisfactory [Henshaw et al, 2012a].

- A non parabolic release rate of steels may indicate a change in the inner oxide phases as plant ages;
- The nickel release or corrosion rate of Alloy 690 falls significantly after cycle 3 or 4;
- The decrease in Co-58 release with time may be explained by the decreasing fraction of Ni metal in crud which is becoming richer in nickel ferrite, less released during shutdown;
- The fitted corrosion rate is  $1.4 \cdot 10^{-4} t^{-0.5}$  (g m<sup>-2</sup> s<sup>-1/2</sup>) for Vandellos II SG and  $0.9 \cdot 10^{-4} t^{-0.5}$  for Sizewell SG.

The life cycle of C-14 has been studied for VVER units for its long half live of 5730 years and capability of integration into the environment [Lysakova et al, 2012]. It is produced on earth by the <sup>14</sup>N(n,p)<sup>14</sup>C reaction and in NPP by the following reactions: <sup>13</sup>C(n,γ)<sup>14</sup>C, <sup>14</sup>N(n,p)<sup>14</sup>C, <sup>15</sup>N(n,d)<sup>14</sup>C, <sup>16</sup>O(n,<sup>3</sup>He)<sup>14</sup>C, <sup>17</sup>O(n,α)<sup>14</sup>C, with the main quantity coming from N-14 having a high thermal cross-section, followed by several order of magnitude from O-17 and then C-13 while N-15 and O-16 are only produced in Fast Breeder Reactors (FBR). It looked amazing for some of the audience attending the conference that the civil nuclear world has to be concerned about its production of C-14, which is only representing 0.02% of the global inventory on earth, as illustrated on Table 2-1.

Table 2-1: Global inventory of C-14 [PBq] in the year 2000, after [Lysakova et al, 2012].

Source	Inventory	Emissions into the environment	Inventory ratio
Production in the upper atmosphere and terrestrial inventories	12 750	1.54	98.34%
Atmospheric testing of nuclear weapons	213	-	1.64%
Nuclear reactors emissions	2.8	0.15	<b>0.02%</b>
<b>TOTAL</b>	<b>12 966</b>		<b>100%</b>

ANT International, 2012

Due to the new policy in energy generation giving the priority to renewable wind and solar sources, Philippsburg (KKP2) and Neckarwestheim (Germany) had to operate with load follow mainly since 2008-2007 respectively. The applied chemistry in the primary coolant thanks to coordinated chemistry with  $pH_{300^{\circ}C}$  7.4, addition of zinc in 2005 and use of EBA in 2008, activity results are satisfactory, as can be seen on Figure 2-9 [Böttcher et al, 2012].

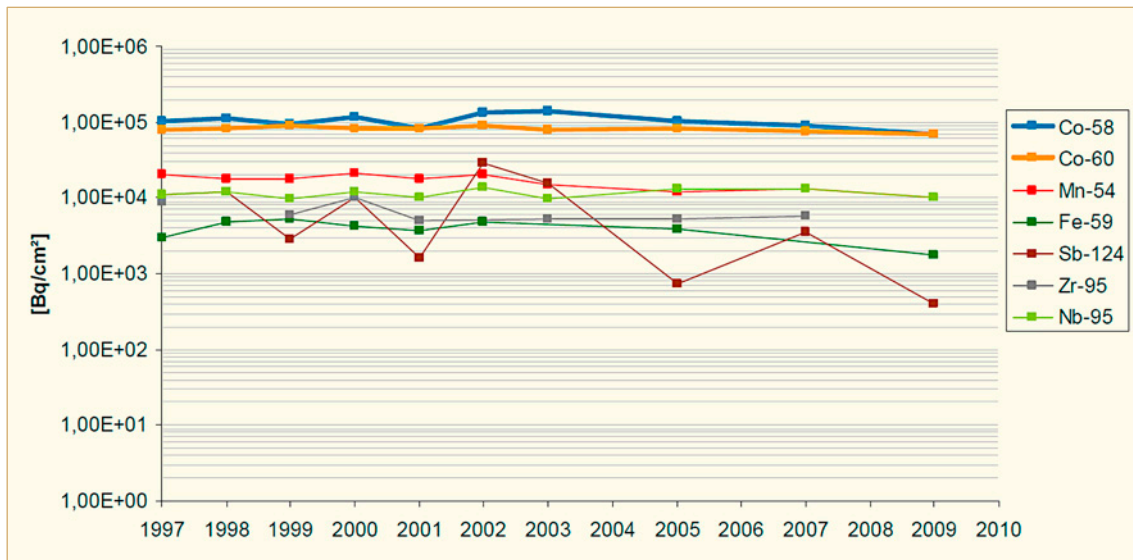


Figure 2-9: Evolution of specific activities on loop 3 at Philippsburg NPP [Böttcher et al, 2012].

A Canadian study showed that the radiolytic degradation of hydrocarbons that may inadvertently enter the RCS would produce hydrogen, with a reaction increasing with the surface area.

## 2.3 BWR primary water chemistry and radiochemistry

### 2.3.1 Treatment options

EPRI [Garcia et al, 2012a] made the first overall presentation of “*Advances in BWR Chemistry*” with the evolution from pure water (highly oxygenated) Normal Water Chemistry (NWC) to Hydrogen Water Chemistry (HWC) in 1983, followed by Noble Metal Chemical Addition (NMCA) in 1995 then On Line Noble Metal application process (OLNC) in 2006, and now Early Hydrogen Water Chemistry (EHWC) addition to always better mitigate SCC of components. It is expected that by 2015, All US BWR plants, sensitive to SCC, will have transitioned to Noble Metals addition.

The IGSCC initiation threshold is -230 mV (SHE) for sensitized austenitic stainless steels and the proportion of operation time where the Electro Chemical Potential (ECP) is  $\leq -230$  mV (SHE) has increased from 93% in 2000 to almost 98% in 2009 thanks to the chemistry improvements [Garcia et al, 2012a].

The idea of EHWC is to inject hydrogen at lower power during start up from refuelling or mid-cycle outages, since a significant percentage of crack growth during an operating fuel cycle is claimed by EPRI to occur during start-up, when reactor temperature is  $> 93^\circ\text{C}$  ( $200^\circ\text{F}$ ) and the concentration of impurities that contribute to Intergranular Stress Corrosion Cracking (IGSCC) are higher than during normal power operation [Garcia et al, 2012a]. In two other papers, EPRI detailed the first industry demonstration carried out at Peach Bottom3 BWR during cycle 19 start up in October 2011 where the hydrogen / oxidant defined as  $\text{H}_2/\text{O}_2 + \text{H}_2\text{O}_2 \geq 2$  has been applied at Reactor Water Cleanup System (RWCU) together with  $\text{H}_2 < 4\%$  at Mechanical Vacuum Pump (MVP) gas + vapour, for a better CGR mitigation on 304 SS.

In a last paper, EPRI focused on sampling and measurement of hydrogen in MVP (no expected sampling in initial design) for applying EHWC chemistry during start-up considering ensuring the limit  $< 4\%$  (lower flammability limit) for hydrogen/air mixtures, the generation of  $\text{H}_2$  and  $\text{O}_2$  from water radiolysis,  $\text{H}_2$  injection and presence of  $\text{O}_2$  from air inleakage [Garcia et al, 2012b]. Figure 2-10 shows the  $\text{H}_2$  content in the gas phase as a function of power for various air inleakages.

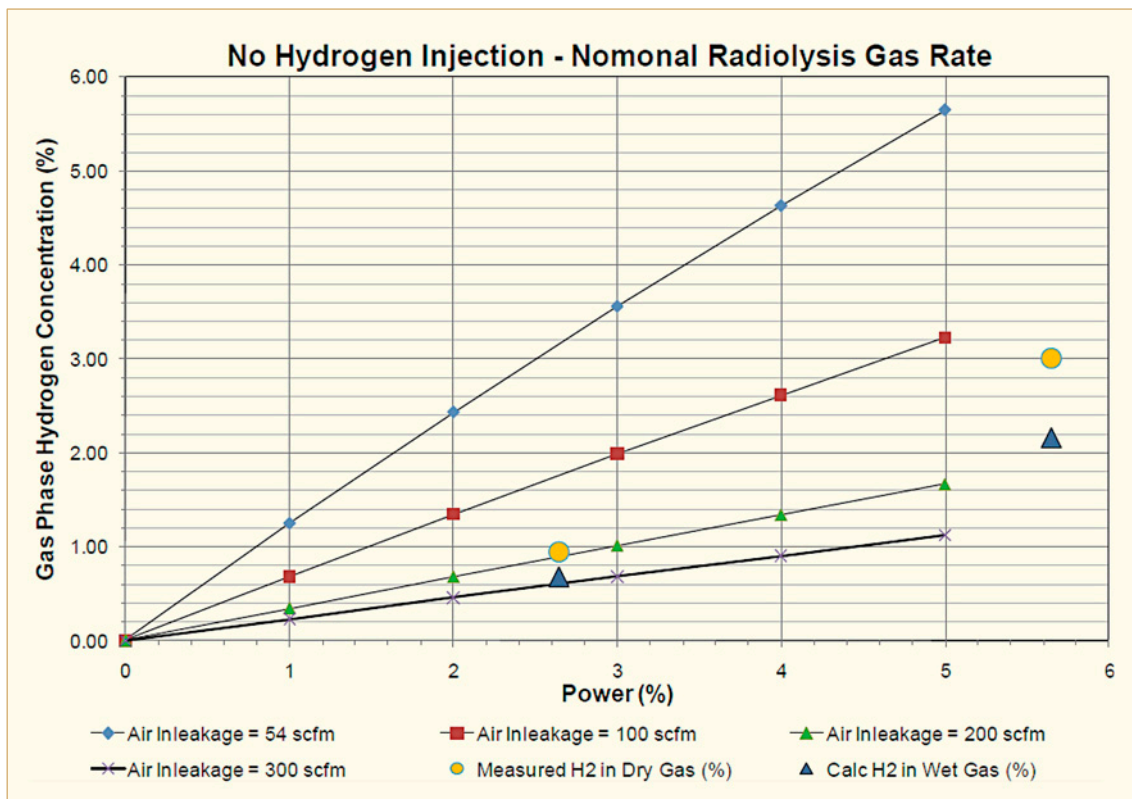


Figure 2-10: MVP H2 (%) in dry gas and calculated in the process (wet gas) [Garcia et al, 2012b].

The proportion and number of units adding hydrogen at various Reactor power shows that many of them are adding hydrogen at rather low power, as shown on Table 2-2.

Table 2-2: Reactor Power at which HWC is placed in service at 35 U.S. BWRs, after [Garcia et al, 2012a].

Reactor Power	< 10%	10 to 25%	25 to 50%	50 to 75%
Number of US BWR Units	16	5	11	3
% of US BWR Units	46%	14%	31%	9%

ANT International, 2012

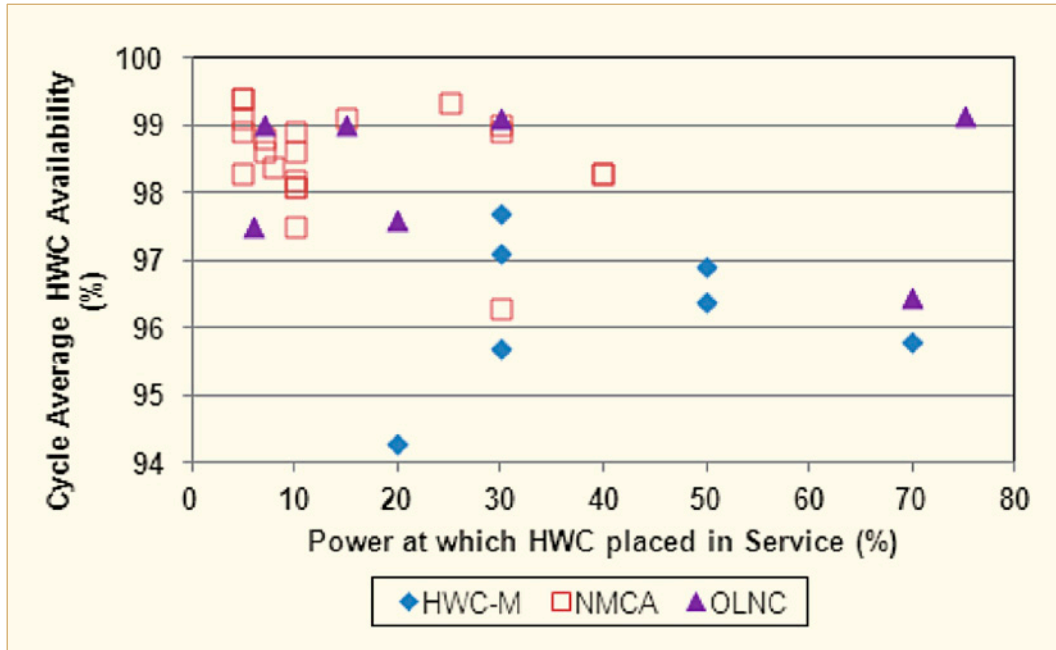


Figure 2-11: Cycle average HWC availability vs. reactor power at which HWC is placed in service at 35 U.S. BWRs [Garcia et al, 2012a].

As it is for PWRs, Radiation field control is an important issue for BWRs, where the BWR Radiation Assessment and Level Control (BRAC) concept has been developed. It clearly shows the benefit of Noble Metal application versus HWC. During NMCA, there is a transformation of the oxide layer with release of Co-60, similar to, but at a lesser degree, than the effect of chemical decontamination [Garcia et al, 2012a].

A great improvement in BRAC values is coming from the addition of zinc in all US BWR units since 2009. Depleted Zinc Oxide (DZO) is injected to minimize the presence of Zn-65 radioisotope.

The important reduction of feed water iron in BWRs is also a key issue and is achieved by the use of filters and optimized use of Ion Exchange Resins (IER) in Condensate Polishing Plant (CPP) and Reactor Water Cleanup Systems. Feed water iron at U.S BWR decreased from an average of 1.2 ppb (µg/kg) in 2000 to 0.44 ppb in 2010 [Garcia et al, 2012a]. As shown on Figure 2-12, this 60% reduction was obtained by

- The installation of pre-filters at plant with only Deep Bed (DB) CPP.
- The increased use of high efficiency iron removal septa at plants with Filter demineralisers (F/D) CPP. BWRs with F/D have transitioned from the use of wound yarn septa filters with high powdered pre-coat dosages to the use of high efficiency pleated septa with lower pre-coat dosages (90 of such units in 2010).



Since the feed water iron concentration decreases, the required concentration of zinc to be added also decreases to keep the target zinc concentration factor ratio for Occupational Radiation Exposure (ORE). The side benefit of a lower zinc concentration is a lower probability of tenacious crud (zinc ferrite) deposition that may reduce fuel performances.

The Reactor Water quality improvement with Condensate Polishing Plant (CPP) strategies also resulted in a better efficiency of Ion Exchange Resins (IER) for anions elimination. It consists in using anions resins under layers to remove cation resin decomposition products and using high cross-linked gel cation resins for improved sulphate control due to improved oxidative stability [Garcia et al, 2012a]. Consequently, from 2000 to 2010, the average chloride and sulphate concentrations decreased, as shown in Table 2-3. The benefit is a lower risk of corrosion that may be enhanced by the presence of anions, which are able to concentrate on heat transfer surfaces.

Table 2-3: Anions concentration improvement in Reactor Water of U.S. BWRs, after [Garcia et al, 2012a].

Parameter	Average concentration in 2000	Average concentration in 2010	% decrease
Chloride	0.40 ppb	0.27 ppb	33%
Sulphate	2 ppb	0.9 ppb	55%

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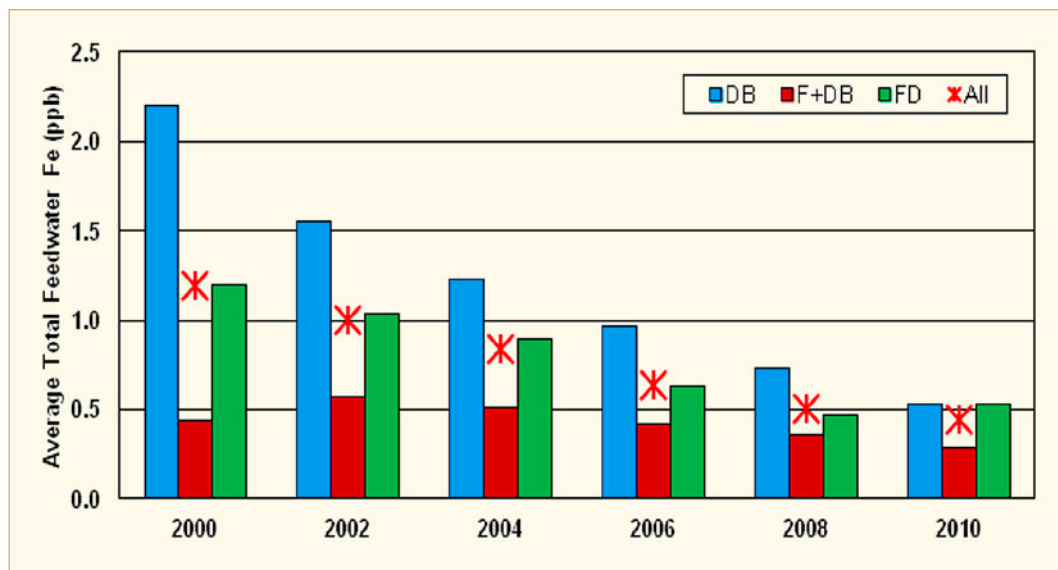


Figure 2-12: U.S. BWR feed water iron evolution from 2000 to 2010 [Garcia et al, 2012a].

A management of hydrogen addition (HWC) has been applied at Kuosheng BWR units in Taiwan for mitigating ORE coming from hydrogen addition, which has been increased from 0.5 to 1 ppm in 2009. Shutdown cooling at lower temperature has been applied, like in Shika-2 (Japan), with inconsistent results between the 2 units. Among the other actions for trying to decrease ORE, a higher coolant intake from the lower plenum at RWCU, an enhanced clean-up of the fuel pool water, use of low Co materials were applied. However, it is difficult to consider that these are real improvements since none of these BWRs with HWC are adding zinc and their BRAC index is much higher than most other BWR units. It has also been mentioned that the government should not give any life extension and that 2026 should be the end of NPP in Taiwan [Tsung-Kuang et al, 2012].

General Electric (GE) described the chemistry changes and monitoring implemented when moving from NMCA to OLNLC for IGSCC mitigation. OLNLC™ is using sodium hexahydroxyplatinate  $\text{Na}_2\text{Pt}(\text{OH})_6$ . The chemistry changes may be encountered in the reactor are conductivity and sodium increase, zinc decrease, iron, filterable activated (Co-60, Co-58, Mn-54, Fe-59, Zn-65 and Cr-51) corrosion by-products increase and to a lesser extent sulphate increase from hide out return. ECP measurements are considered to be a better surface mitigation than noble metal loading to demonstrate Pt deposition, which is the advantage of ONLC over NMCA. The other advantages are better penetration of Pt in existing cracks, reduction in time period between reapplication, reduction in chemistry transients, elimination of the impact on the duration of refuelling outages [Varela et al, 2012].

A Swiss Institute investigated Pt deposition behaviour in a high-temperature loop under BWR HWC and NWC conditions. With an excess of  $\text{H}_2$ , fine Pt particles should have a better catalytic efficiency while an excess of  $\text{O}_2$  gave larger particles. Consequently, a reducing environment is favourable for a more effective deposition, with lower ECP and better protection against SCC. A longer pre-oxidation seems to increase the Pt concentration at the specimen surface and is also beneficial. In another paper, the Institute claimed that a lower Pt injection rate is inducing smaller Pt particles, favourable for SCC mitigation.

AREVA demonstrated the effectiveness of methanol (MeOH) for preventing RPV internals from SCC in locations where cold worked materials are exposed to oxygenated high temperature water. During tests under irradiation by vacuum UV at  $125^\circ\text{C}$  (similar to Cherenkov radiation), MeOH is able to shift the corrosion potential from oxidizing to reductive conditions ( $< -230 \text{ mV}_{\text{SHE}}$  for austenitic stainless steel) with MeOH /DO ratio  $\geq 1$  as illustrated on Figure 2-13 [Stellwag et al, 2012a].

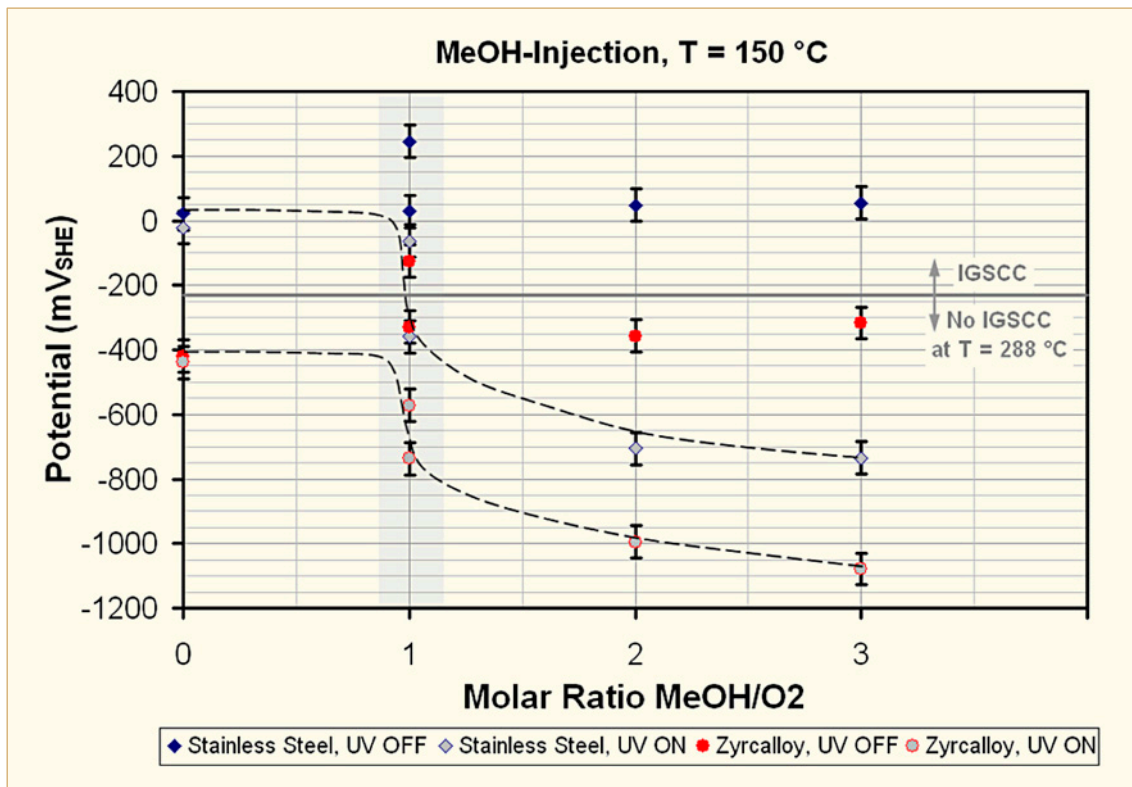


Figure 2-13: Corrosion potential of stainless steel 347 and Zry-4 as a function of molar ratio MeOH/O<sub>2</sub> at 150°C with O<sub>2</sub> of 200 to 250 ppb, MeOH = 0, 200, 400, and 600 ppb, electrical conductivity 0.06  $\mu\text{s}/\text{cm}$  [Stellwag et al, 2012a].

Another paper from AREVA reported tests on the decomposition of methyl iodide associated to methanol application in BWRs. Based on the tests, it is concluded that using methanol < 2.5 ppm for IGSCC mitigation of internals is inconsequential to the accident conditions related to methyl iodide formation.

The EPRI model code for radiolysis on BWR has been applied and compared to plant data for providing an accurate molar ratio of hydrogen / oxidant defined as  $[H_2]/[O_2+0.47[H_2O_2]]$  in various situations, including plants adding Pt to catalyze hydrogen addition. It appears that a ratio >2 is necessary to get reducing conditions and thus a protection against SCC in presence of Pt. ECP measurements show different responses from plant to plant upon hydrogen concentration to achieve the required ECP < -230 mV [Henshaw et al, 2012b]. Figure 2-14 shows comparison of prediction versus measured values of the molar ratio in the recirculation lines of 3 BWR plants.

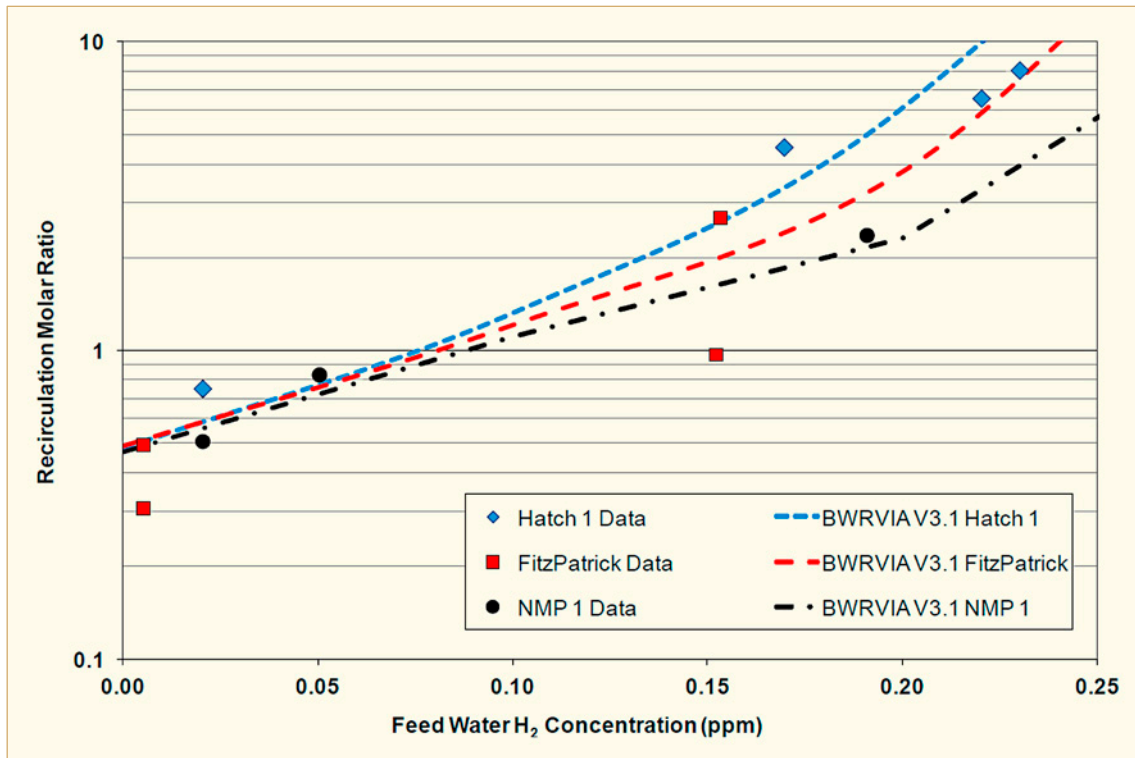


Figure 2-14: Comparison of measured versus model prediction of the hydrogen / oxidant molar ratio in 3 BWR units [Henshaw et al, 2012b].

In another poster paper, the ratio  $H_2/(O_2+H_2O_2) > 2$  is mentioned to be necessary to get an ECP ~ -500 MV on noble metal treated surface and consequently being in a range of IGSCC mitigation [Cowan & Garcia, 2012]. The author considers that Zn presence at a concentration of 100 x that of soluble cobalt could significantly reduce Co uptake in the corrosion film and consequently lower the BRAC dose rates than BWR operating under HWC without zinc, although the benefit of zinc is not always so important since not all the plants operate with ECP near H<sub>2</sub> redox potential. The advantage of NMCA is to establish low ECP and thus protection against IGSCC, while keeping low BRAC values if the ratio  $^{60}Co (s)/Zn (s) < 0.74$  Bq/ml as illustrated on Figure 2-15 and Figure 2-16. However, BRAC values may increase in following cycles with the lost of Pt and new film structure incorporating more Co. OLNc allow to keep low BRAC values even during the following cycles or to reach them after some fuel cycles, see Figure 2-17. OLNc is able to repair the local Pt effectiveness. The way of maintaining low BRAC for the three IGSCC mitigation methods is to keep low ECP in recirculation system at - 500mV SHE while maintaining Zn injection on feed water. Finally, ratio  $^{60}Co (s)/Zn (s) < 0.74$  Bq/ml during and after OLNc applications should get low BRAC dose rates < 1.5mSv/h and even lower after several cycles [Cowan & Garcia, 2012].

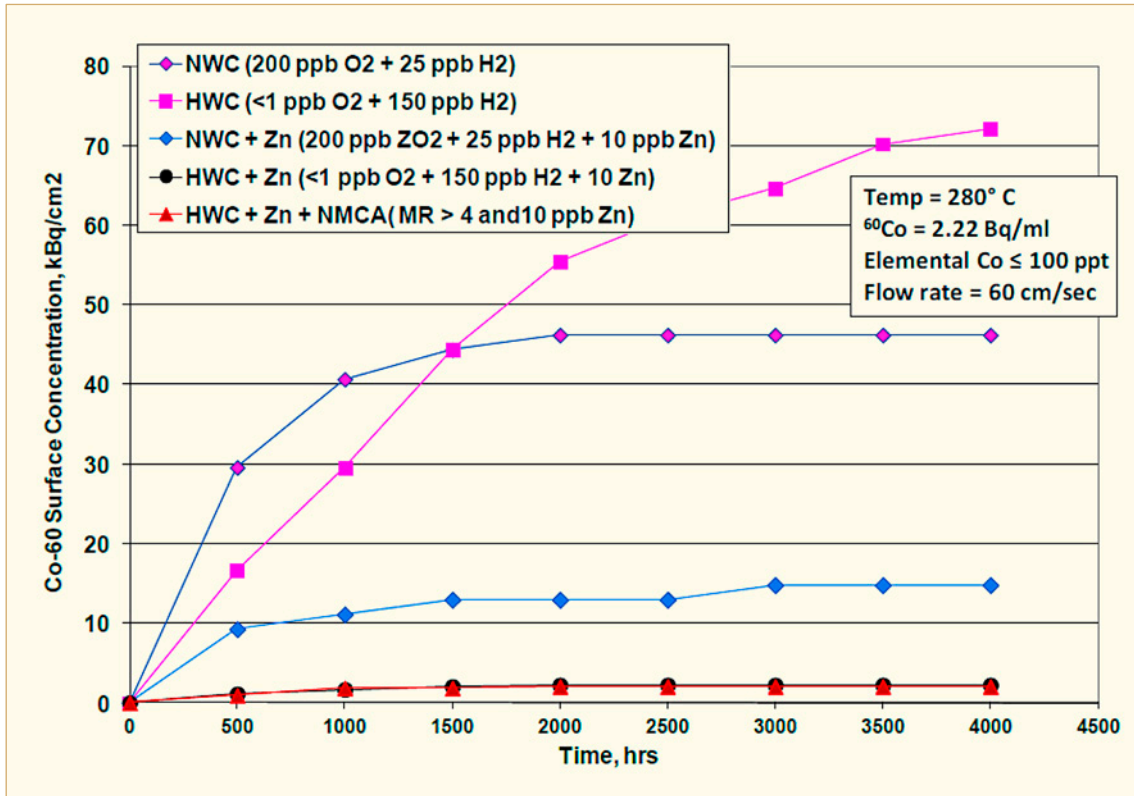


Figure 2-15: Effect of different BWR environmental conditions on Co-60 incorporation into the corrosion film on 304 stainless steel [Cowan & Garcia, 2012].

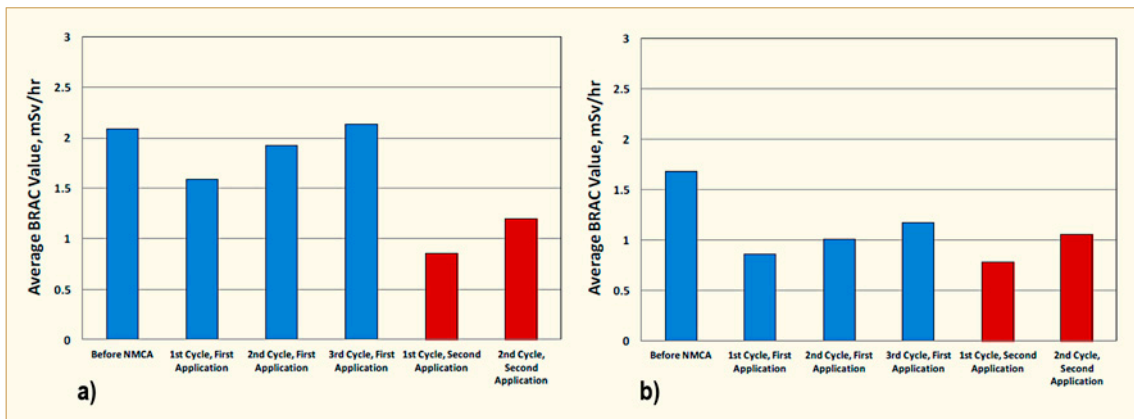


Figure 2-16: BRAC values before and after NMCA applications (a) all treated BWR and (b) BWRs that had  $^{60}\text{Co}/\text{Zn(s)}$  ratios  $< 0.74$  Bq/ml/ppb in the cycle immediately after NMCA application [Cowan & Garcia, 2012].

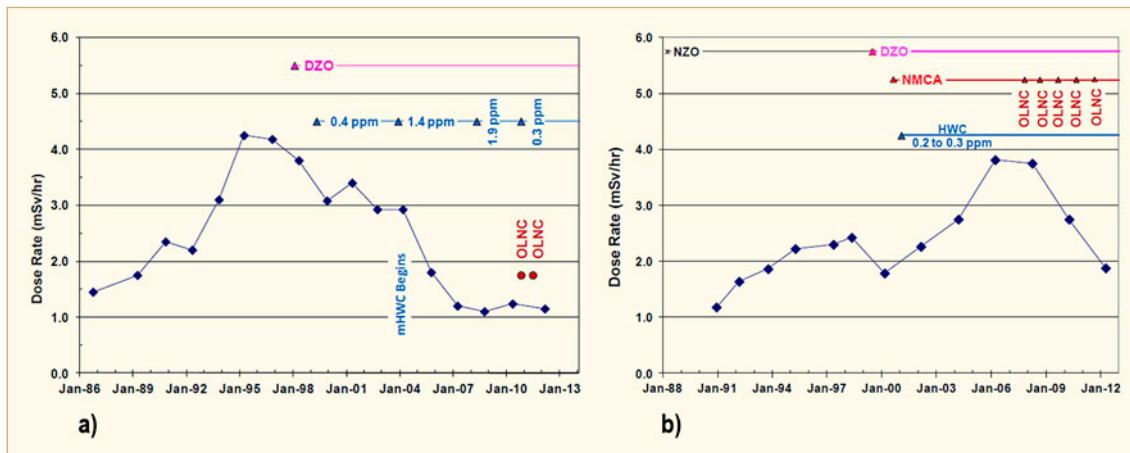


Figure 2-17: OLNLC response at two BWRs (a) mHWC to OLNLC and (b) NMCA to OLNLC [Cowan & Garcia, 2012].

For safety improvements, a computer code has been developed by a University in Taiwan for qualitatively predicting the coolant chemistry of ABWR, namely the variation in redox species, radiolysis effect and concentrations of  $H_2O_2$  and  $O_2$  which may become high inside or near the core.

## 2.3.2 Dose rate and zinc

An experimental system has been built at Studsvik (Sweden) to study activity build up in BWR as a function of chemistry with the effect of combined addition of Zn and Fe. The comparison of activity build up for various ratios of Fe (ppb) / Zn (ppb) shows that a ratio 2 ppb Fe / 0.5 ppb Zn is sufficient to induce a decrease of Co-60. When Zn increases to 5 and 10 ppb, the radioactivity build up is reduced and a ratio of 2 ppb Fe / 5 ppb Zn almost stops the activity build-up. Zinc incorporates only into the inner oxide layer with a competitive process between Zn and Co; consequently, if Zn is increased, there will be less Co incorporated in the constant thickness of the oxide layer and thus less radioactivity deposition. Outer and inner oxide layers are respectively about 420 and 120 nm thick, and composed of sub-micron grain of  $NiFe_2O_4$ ,  $(Fe,Cr)_2O_3$  or  $(Fe,Cr)_3O_4$  for the outer layer and mixed oxide spinel phase of  $Ni_{0.3}Fe_{1.5}Cr_{1.2}O_4$  for the inner layer [Gustafsson et al, 2012].

Under the BRAC indicator, it has been observed that when adopting HWC for SCC, dose rates increased, which has been at the origin of the decision to move to NMCA and then OLNLC™ and also to add zinc, all these chemistries allowing to decrease again dose rates. Japanese made experiments to evaluate the respective effect of Pt and Zn on Co-60 deposition behaviour. The tests showed that the presence of Pt on a surface causes a reduction of ~50% in Co deposition. Hydrogen peroxide, even at low concentration (< 5ppb) had a tendency to accelerate Co-60 deposition. The combination of chemical decontamination followed by short Pt deposition and Zn injection seems an effective method to reduce BRAC dose rate under NMCA [Nagase et al, 2012].

A Taiwanese Institute verified that zinc addition in BWR did not affect significantly the surface oxides of SS 304 and thus the SCC mitigating effect of HWC. With zinc addition, a substantial cathodic shift is observed under UV illumination, meaning an additional protection of in-core components when HWC is not in operation.

In order to decrease Co-58 and Co-60 source of radiation in Lugmen ABWR (Taiwan), a pre-treatment was applied in 2010 for passivation the metal surfaces. The passivation was achieved by pre-filming on piping before operation, at 180-230°C and pH 8.0-8.5 with NaOH injection.

### 3 Secondary water chemistry (NPC 2012) (Francis Nordmann)

#### 3.1 General information

Dow Chemical presented a new resin to mitigate the anion kinetic impairment issue with PWRs using ethanolamine (ETA). The test was done at Diablo Canyon (USA) and the resin properties did not change after 1.5 years of service. This is of great importance with the increasing use for many reasons of ETA as a conditioning amine reagent for the secondary system of PWR units.

Steam Quality for various elements (Ca, Mg, K, Cu has been determined in Argentina with satisfactory results.

EDF prepared an optimized strategy for chemistry during start up so as to save time, quantity of used water and generated wastes coping with a sufficient chemistry quality. Practical recommendations are given. In another paper related to the same objective, EDF is focusing on the use of Mobile purification systems that are used in French PWR units for cleaning the secondary system, treating the feed water. The principle is to use mobile purification systems of limited flow, with non regenerated IER, used only during start-up, avoiding traditional condensate polishing plants for saving investment and operating costs, decreasing the risk associated with IER regeneration and other objectives that have been fully covered in previous documents [Nordmann, 2006]. This suitable strategy is also applied for the EPR and is now extended at all the French fleet, including the plants where such a system had not been installed in the late 1990s as one of the remedy for mitigating secondary side IGA/SCC of SG tubing with Alloy 600 on river water.

#### 3.2 Secondary system treatment strategy

With the evolution of degradation risks on the secondary system in Japanese PWRs, the focus has changed. During the 3 last decades, the SG troubles decreased respectively from 34 to 19 and 0 for OD IGA/SCC which is not any more an issue with the SG with sensitive Alloy 600 MA that had SGR. There are now 18 PWR units with 690 TT and 6 with 600 TT SG tubing. The progressive elimination of copper alloys also allowed a pH increase, with the well known benefit of mitigating FAC of carbon steel. Moreover, the extensive use of Stainless steel in replacement of copper alloys in LP-and HP heaters and in MSR is also a key way of avoiding FAC. Thanks to the exclusive use of Ti for condenser tubing, their tightness is now allowing to by-pass the condensate polishers which is favourable to increase the pH in the secondary system. These CPP are only used during start up and shutdown. Either ethanolamine (ETA) with a pH of 9.8 in feed water or H-AVT with ammonia at a pH > 9.8 is recommended. Such improvements allowed to drastically decrease the iron concentration in feed water which decreased for example from about 6 to 1 ppb at Genkai 1 and to  $\leq 1$  ppb at Tomari 3. As it can be seen on Figure 3-1, there were in 2010, respectively 6 units with AVT (NH<sub>3</sub>), 10 with ETA, 4 with H-AVT (high pH with NH<sub>3</sub>) including 1 with some oxygen addition, 4 with High ETA (high pH with ETA) [Maeda et al, 2012].

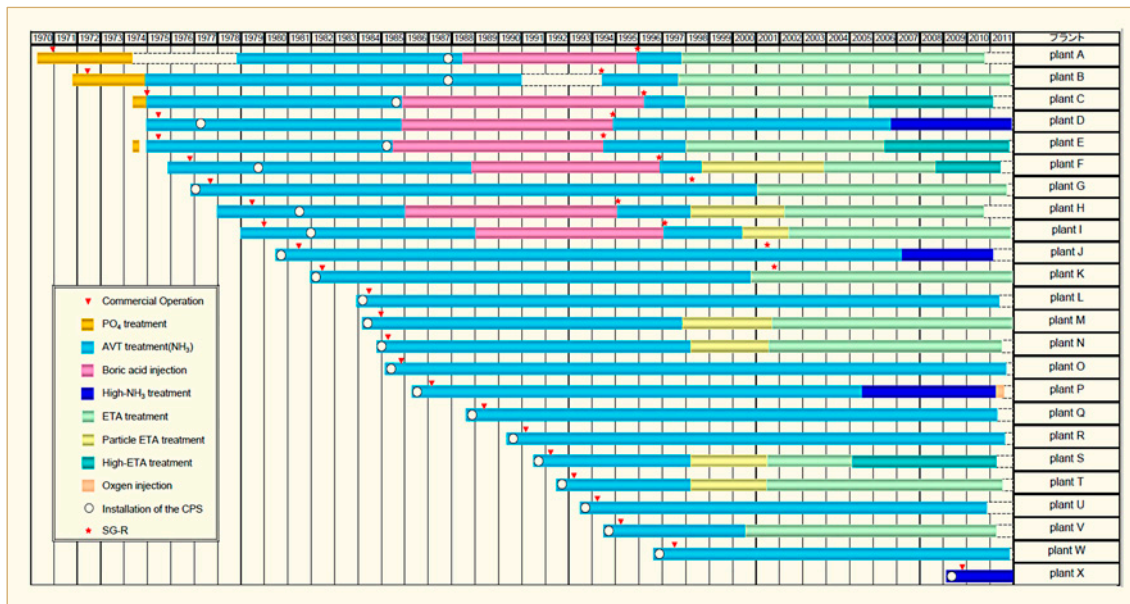


Figure 3-1: History of Secondary system Treatment in Japanese PWR units [Maeda et al, 2012].

ANT-International presented an overall paper of strategy for the secondary side issues and remedies in PWR and VVER Plants. The conclusions are the following [Nordmann et al, 2012]:

Secondary side degradation of steam generators (SG) tubing with Alloy 600 MA and Flow Accelerated Corrosion (FAC) of Carbon Steel have been for a long time important issues for the secondary system of PWR and VVER. With the beneficial evolution of the design, the most important issues are progressively moving to a larger variety of risks associated to potential inadequate chemistries.

FAC may be easily mitigated by selecting components with sufficient chromium content. In addition, an appropriate treatment with ammonia at a  $\text{pH}_{25^{\circ}\text{C}} > 9.8$  or by amine treatment will be also a clever option. At the same time, these remedies will minimize corrosion products transport toward the SG that will have several positive effects:

- Mitigating the risk of Flow Induced Vibration;
- Minimizing the risk of concentration of impurities in local areas where they may induce corrosion;
- Decreasing the need for maintenance cleaning activities or for the need to add dispersant;
- Keeping a high heat flux and plant performances.

The various remedies for having clean steam generators, with low quantities of corrosion products includes an optimum treatment, maintenance cleaning activities (either curative hard chemical cleaning or the interesting option of Advanced Scale Conditioning Agent (ASCA) application or the efficient addition of dispersant in the feed water, which drastically increases the efficiency of corrosion products elimination through SG blow down.

The influence of impurities depends on the SG tubing material. It is mandatory to completely avoid alkaline environment (sodium) for Alloy 600, and to avoid acidic chlorides for Russian alloy 18-10 Ti. With Alloy 690 or 800, it remains important to avoid severe deviations (highly alkaline, or acidic, lead or resin fines pollutions and more generally reduced sulphur species).

The need for very low oxygen levels is not any more a good approach and it may even be interesting to keep a few  $\mu\text{g}/\text{kg}$  (ppb) at the condenser level but still having a reducing environment in the final feed water and in the SG.

The new challenges should not only focus on reaching a very pure chemistry, when this is not necessary, but will also concentrate on selecting the best SG cleaning methods, on minimizing liquid and solid wastes. A good example is the absence of having condensate polishers in operation for the plants with tight condensers.

In the same way, several improvements may be implemented in optimizing the number of on-line monitors and chemical controls to what is of some use and eliminating regular controls when occasional diagnosis is sufficient [Nordmann et al, 2012].

An AREVA presentation gave some similar points of view on secondary chemistry objectives, i.e. the decrease of corrosion products through a sufficient pH in various parts of the system. However, preference is given by the German part of AREVA to the use of only ammonia at a pH  $\geq 9.8$ , produced by hydrazine, since it makes the treatment simpler and avoids some drawbacks of amines. The target  $N_2H_4$  concentration is 100  $\mu\text{g}/\text{kg}$  and a  $\text{pH}_{180^\circ\text{C}} > 6.6$  in wet steam phase, which is the temperature with the highest FAC rate. CHEMBAL code is used to perform detailed mass balance calculations and adjust the treatment strategy to a plant specific cases, depending on the component to be protected against FAC with 3 examples of strategies with the respective added reagents  $N_2H_4$  (100 ppb) +  $NH_3$  or  $N_2H_4$  + ETA or  $N_2H_4$  +  $NH_3$  + ETA [Roumiguière et al, 2012].

### 3.3 Corrosion products and FAC in secondary system

As explained in the section related to strategy of secondary system treatment, Oxygenated Water Chemistry (OWC) consists in the addition of small quantities of dissolved oxygen (DO) in the parts sensitive to FAC is one of the efficient remedies to mitigate the phenomenon, particularly if other ones cannot be sufficiently implemented (pH increase, material selection). The Japan Atomic Power Co. (JAPC) carried out tests at Tsuruga 2 demonstrating the efficiency of such oxygen addition to mitigate FAC, thanks to continuous monitoring by high-temperature and high-resolution ultrasonic probe [Sugino et al, 2012]. As highlighted in Table 3-1, APC concluded that FAC is respectively mitigated by 5 ppb of DO under low AVT (pH 9.3) and 2 ppb under High AVT pH 9.8). Such addition has no adverse effect since. This has been crosschecked by Electro-chemical Potential (ECP) measurements showing that ECP necessary to get stable hematite respectively correspond to 5 ppb of DO under low AVT treatment and 2 ppb under High AVT, as shown on Figure 3-2.



## 4 Auxiliary systems, cooling water systems, water and waste (NPC 2012) (Francis Nordmann)

### 4.1 Fukushima treatment

Two presentations related to the treatment of Fukushima event have been done. The first special of TEPCO explained how the contaminated water has been treated for with desalination by reverse osmosis and evaporation concentration. To remove radioactive materials two caesium adsorption equipments and one decontamination by flocculation have been implemented. Desalination decreased chloride from 2300 down to 7 ppm and evaporation from 6900 to 2 ppm. There have been 3 steps: elimination of Cs 134 & 137; elimination of chloride and purification on resins [Makihira et al, 2012].

The second presentation, from Veolia, a company in charge of water treatment was also related to the treatment of contaminated water jointly designed with AREVA. The purpose was too quickly achieve a stable situation of the reactors, allowing treated water to be reused for core cooling. Micro-sand with lamellar setting Actiflo™ has been used.

### 4.2 Waste treatment

A Low Level active liquid Waste (LLW) purification system has been installed at Bruce NPP (Canada). It includes three sub-systems

- A pre-treatment to remove suspended solids and colloidal particles;
- A high recovery 3 stage membrane system achieving > 99% combined permeate recovery, with Reverse Osmosis, producing a pure permeate free of chemical and radioactive contaminants, except tritium; it is followed by ultra-filtration membrane;
- A wiped film down-flow evaporator to treat the small membrane system reject stream.

The main Indian Laboratory experimented the electro-deposition of metal ion (mainly Fe<sup>II</sup>) by suitable voltage to treat the wastes from decontamination.

French CEA Laboratory developed a 3-steps chemical separation procedure to isolate <sup>129</sup>I, a fission product from <sup>235</sup>U and <sup>239</sup>Pu, for its analytical determination by accelerator mass spectrometry. This is necessary to measure very low values of long-life isotopes in radioactive waste management.

An Austrian abstract is explaining the interest to store zeolite in the unlikely case of emergency situation where it would be necessary to adsorb nuclear radiation from radioactive elements thorough their cation exchange properties. Zeolite has been successfully used in Chernobyl and Fukushima for nuclear wastes repository. Zeolite with high content of clinoptilolite (>90%) have a binding capacity of 2.5meq/g for caesium and 1.4 meq/g for strontium ions.

Hydrazine being a carcinogenic compound, its use and even more its discharge is highly controlled. Hydrazine injected in the secondary system (feed water) during power operation is:

- Either reacting with oxygen  $N_2H_4 + H_2O + O_2 \rightarrow N_2 + 3 H_2O$
- Or thermally decomposed  $N_2H_4 \rightarrow N_2 + 2H_2$

However, during lay-up of SG, the large quantities of remaining hydrazine are not thermally decomposed and it is important to eliminate it before discharge to the environment. The Belgian Laboratory (Laborelec) studied various methods for this purpose of hydrazine decomposition, using various oxidants and catalysts [Schildermans et al, 2012]:

- Bleach,  $\text{N}_2\text{H}_4 + 2 \text{NaOCl} \rightarrow 2 \text{NaCl} + \text{N}_2 + 4 \text{H}_2\text{O}$  (may also produce chlorine gas)
- Hydrogen peroxide  $\text{N}_2\text{H}_4 + 2 \text{H}_2\text{O}_2 \rightarrow \text{N}_2 + 4 \text{H}_2\text{O}$  with catalysts, potassium permanganate ( $\text{KMnO}_4$ ), copper sulphate ( $\text{CuSO}_4$ )

The numerous tests have shown the influence of:

- Temperature with a positive effect on kinetics, faster at 25 than 5°C;
- The added catalyst;  $\text{KMnO}_4$  and  $\text{CuSO}_4$  both have an optimum concentration of 10 ppm;
- The oxidant, with an optimum stoichiometric ratio oxidant / hydrazine of 1 - 1.5;
- The advantage of  $\text{CuSO}_4$  over  $\text{KMnO}_4$  because the neutralization with  $\text{CuSO}_4$  is faster and sulphate from it may be marginal as compared to sulphuric acid used for cooling system treatment;
- A degradation in laboratory condition completed in < 1.5 hour for  $\text{H}_2\text{O}_2$  vs. a few minutes with  $\text{NaOCl}$  (see Figure 4-1) but an expected higher duration in SG conditions (impurities, imperfect mixing).

When using  $\text{NaOCl}$  for eliminating 75 ppm  $\text{N}_2\text{H}_4$  from a SG in lay-up condition containing 20 ppm ETA, a maximum of 1.8 kg (1.5 mg/l) of volatile organic compounds is transferred into the atmosphere and ~ 225 g of absorbable compounds remain in the neutralized solution. Finally, it is concluded that  $\text{NaClO}$  and  $\text{H}_2\text{O}_2$  each have their own advantages and disadvantages.  $\text{NaClO}$  is faster and cheaper, does not need catalyst but is producing some undesirable compounds released (particularly in presence of ETA) while  $\text{H}_2\text{O}_2$  does not produce such undesirable compounds [Schildermans et al, 2012].

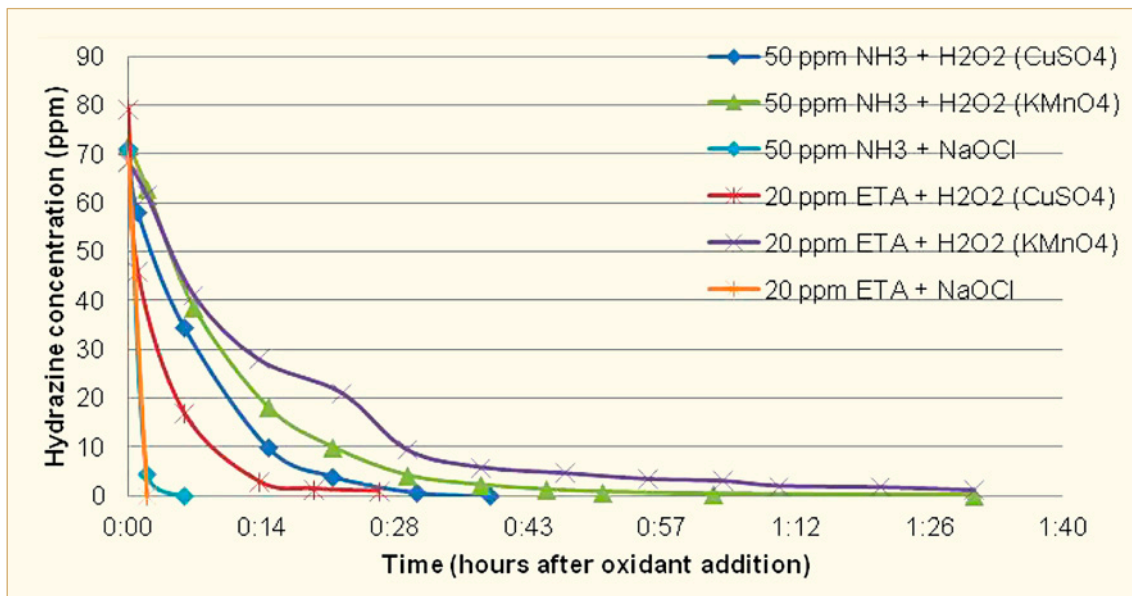


Figure 4-1: Hydrazine degradation kinetics in SG lay-up solutions with ETA and NH<sub>3</sub> [Schildermans et al, 2012].

EDF studied alternate compounds in replacement of hydrazine due to its carcinogenic properties and more and more strict regulation with REACH legislation, in force since June 2007. Information has been provided by the end of 2008, as per REACH deadline. Hydrazine is a carcinogenic substance of category 1B, imported in quantity exceeding 1 ton per year, and thus requiring a number of specific rules detailed in the paper [Spahic et al, 2012]. The absence of suitable alternate compounds, able to meet the technical requirements entails rigorous arrangements to be implemented in order to segregate the zones where use of hydrazine is made and therefore eradicate the risk to personnel regarding the hydrazine effect. Consequently, a number of engineering changes and modifications are to be carried out on the chemical injections systems of the French fleet of 58 PWR units over the next years. It is still a pending issue to know whether or not hydrazine will be authorized in the future under REACH regulation, which, for the moment, only gave a temporary derogation for its use.

EDF had to look for alternate compounds with adequate properties: reductive capabilities, sufficient thermal stability on various conditions (normal power operation in feed water and SG lay-up), corrosion risk compatibility for the product and its decomposition products, volatility, analytical capabilities impact on chemistry (cation conductivity and pH), toxicity and waste.

The list of studied alternate chemicals of Table 4-1 demonstrated that no other chemical is suitable to replace hydrazine and consequently, EDF implemented the modifications and various requirements for mitigating any risk for workers, to continue using hydrazine in its fleet [Spahic et al, 2012].

Table 4-1: List of potential hydrazine substitutes, after [Spahic et al, 2012].

Chemical	Characteristics	Comments
N,N diamino-urea (carbohydrazide)	1C Organic compound. Decomposes into N <sub>2</sub> H <sub>4</sub> . Good efficiency at low temp (lay-up). Lower efficiency than N <sub>2</sub> H <sub>4</sub> at high temperature	Toxic by decomposition (N <sub>2</sub> H <sub>4</sub> ) impact on conductivity and acidification (CO <sub>2</sub> )
N, N Diethylhydroxylamine (DEHA)	Organic compound	Produces organic acids impact on conductivity potential corrosion risk
Isopropylhydroxylamine (IPHA)	Organic compound Lower efficiency than N <sub>2</sub> H <sub>4</sub>	Produces organic acids impact on conductivity potential corrosion risk
Methylethylcetoxime (MEKO)	Organic compound Lower efficiency than N <sub>2</sub> H <sub>4</sub>	Toxicity CMR C3. Produces organic acids. Impact on conductivity. Potential corrosion risk
1-Amino-4-methylpiperadine (1A4MP)	Organic compound long carbon chain	Doubt on toxicity Decomposition products? Potential corrosion risk
Erythorbic acid	Organic compound	Produces organic acids impact on conductivity potential corrosion risk
Hydroquinone	Organic compound	Toxic. Previously a catalyst of N <sub>2</sub> H <sub>4</sub> for low temp, now forbidden
Ascorbic acid	Organic compound. Good efficiency at low temperature (wet lay-up)	Low stability at 260°C. Impact on pH and cation conductivity
Octadecylamine (ODA)	Organic compound filming amine (thin layered deposit)	Can be used during lay-up, but removed before power operation

ANT International, 2012

NUKEM company briefly described the possibility of pyrolysis for wastes elimination, including IER.

## 5 Lifetime management and plant ageing (NPC 2012) (Francis Nordmann)

EDF decided to upgrade its chemistry specifications, for improving chemistry management for a long term operation of its fleet in a safe and efficient way. The French specifications have been compared to EPRI (USA) and VGB (Germany) guidelines and it has been decided to have 42% without any change, 25% with a quick decision and 33% deserving some studies. For make-up water, which has a strong impact on the chemistry quality of the various systems, there will be a decision in 2014 whether there will be a new design / revamping or a heavy maintenance of existing stations. The chemistry modifications will include an increased monitoring frequency of some parameters and a focus on chemistry indicators including primary and secondary water chemistry and thus mixing chemistry and radiochemistry, while the previous WANO index was only looking at secondary water chemistry at SG blow down [Bretelle & Stutzmann, 2012]. A poster paper gave additional details on the 2011-2016 project for improving EDF Chemistry specifications with International Benchmarking (EPRI, VGB), which induced 300 recommendations.

On the primary coolant, EDF explained the reasons for keeping a target pH<sub>300°C</sub> of 7.2 and not higher as in many countries with nickel base alloys: avoiding any risk with Li > 3.5 ppm for SCC of stainless steels, absence of authorization from the Regulators to operate at higher Li at Beginning Of Cycle (BOC) that might induce fuel cladding corrosion until changing for M5 or Zirlo, taking into account the operating in various modes, such as start up transients, marginal impact of a pH increase from 7.2 to 7.4 as compared to other parameters for ORE [Bretelle & Stutzmann, 2012].

In the context of Plant Life Time extension, all SG with alloy 600 MA (5 remaining units) or 600 TT (27 units) will be replaced by SG with Alloy 690 as already done in 20 units or in original SG of the 6 more recent units, thus avoiding the PWSCC issue. There is still the concern of Bottom Mounted Instrumentation Nozzles (BMI) made of Alloy 600 with Alloy 182 welding, and a few bimetallic welds in Alloy 82, sensitive to PWSCC, explaining the decision to operate in the upper DH range (see Section 2.7.1).

After the high consequences of FIV of SG tubes in several EDF units, due to a specific unfavourable local design, i.e. absence of Anti-vibration Bar (AVB) support, holes without tubes in broached TSP and important deposits of corrosion products, it appears necessary to decrease the corrosion product transport particularly in units with a low pH that is the consequence of the presence of copper alloys. The objective is to eliminate copper alloys that were present in 15 units cooled by river water, either historically or for counteracting amoebae development due to condenser cooling system having cooling towers. The plants that were operating with ammonia at pH of 9.7 switched to ETA treatment for MSR damages [Bretelle & Stutzmann, 2012]. There is also some pH limitation for environmental constraints associated with nitrogen compounds releases. Including the FAC issue, the optimum treatment is 4 ppm ETA + 1.5 ppm ammonia. For eliminating the deposits in the operating units, a huge program of SG chemical cleanings is implemented and will continue in the 2013-2015 time frame. Finally, carbon steel replacement of reheaters by stainless steel and dispersant addition trial will be implemented to mitigate corrosion product deposition within SG [Bretelle & Stutzmann, 2012].

The Canadian Nuclear Safety Commission presented its oversight to assess the performance of the chemistry program at the 5 NPP with 22 CANDU Reactors in operation at Pickering A1&4 B5-6-7-8; Bruce A1-2-3-4, B5-6-7-8; Darlington A1-2-3-4; Gentilly2 and Pt. Lepreau. A focus has been placed on the Chemistry Index.

## 6 Maintenance activities (NPC 2012) (Francis Nordmann)

### 6.1 Curative cleaning methods - Decontamination

A Full System Decontamination (FSD) with the AREVA process HP/CORD ® UV has been applied at the PWR of Grafenrheinfeld, followed by a concept for sustainable dose rate reduction, with a passivation and high pH operation with zinc addition. The decision to perform a FSD was due to the presence of a high accumulated activity, and this gave a decontamination, factor of ~60 an ORE saving of ~ 4,8 Sv in subsequent outages 2010 and 2011. The dose rates, dominated by Co radioisotope (Co-60 before FSD and Co-58 after FSD) decreased by a factor of 3 after FSD since recontamination, the main issue, has been avoided by zinc injection [Stellwag et al, 2012b].

AREVA detailed the overall strategy for sustainable dose rate reduction which is plant specific and includes: modification of RCS chemistry, elimination of cobalt base materials, outage optimization, permanent shielding, FSD as described just above, formation of a new and very stable protective oxide on the surfaces which may include zinc addition.

The CALLISTO PWR experimental facility in Belgium has been decontaminated to decrease ORE associated to its maintenance and use. 10 GBq of Co-60 have been eliminated with a soft typical process of  $\text{KMnO}_4$  and  $\text{H}_2\text{C}_2\text{O}_4$  allowing a decrease of dose rates by a factor ranging from 2 to 12.

DCNS applied with CEA a new cleaning process using foam for cleaning Alloy 800 heat exchanger (SG) of a naval application where an abnormal chemistry from an excessive corrosion of copper alloys with deposits of copper, iron, nickel phosphates and oxides grown on tubes and tube-sheet had been observed. This naval reactor is applying on the secondary system a phosphate treatment with a mixing of  $\text{Na}_2\text{HPO}_4$  +  $\text{Na}_3\text{PO}_4$ , with a molar ratio  $2.3 < \text{Na}/\text{PO}_4 < 2.6$  and  $9.5 < \text{pH} < 10.2$ . The cleaning objective was to avoid corrosion of SG tubes and restore NDE capabilities. The foam process where the aqueous phase is only 17% allows decreasing the quantity of chemicals and the nuclear waste volume. The cleaning products consist of an aqueous solution with mixing of phosphoric acid (oxide dissolution) + corrosion inhibitor, oxalic acid (iron oxide elimination), hydrogen peroxide (for iron oxalate), surfactants and foam stabilizer. It dissolved 8.8 kg of Cu, 7.3 kg of Fe, 1 kg of Ni and 30 g of Cr in 4 weeks with an electrochemical monitoring to verify that the polarization resistance was remaining higher than a value corresponding to a corrosion intensity of  $50\mu\text{A}$ , and a corrosion rate of 0.5 mm/year. The SG were successfully cleaned, as it can be seen on Figure 6-1 confirming such a procedure could be applied to other cases by adapting the chemicals to the compositions of the deposits [Corrieu et al, 2012].

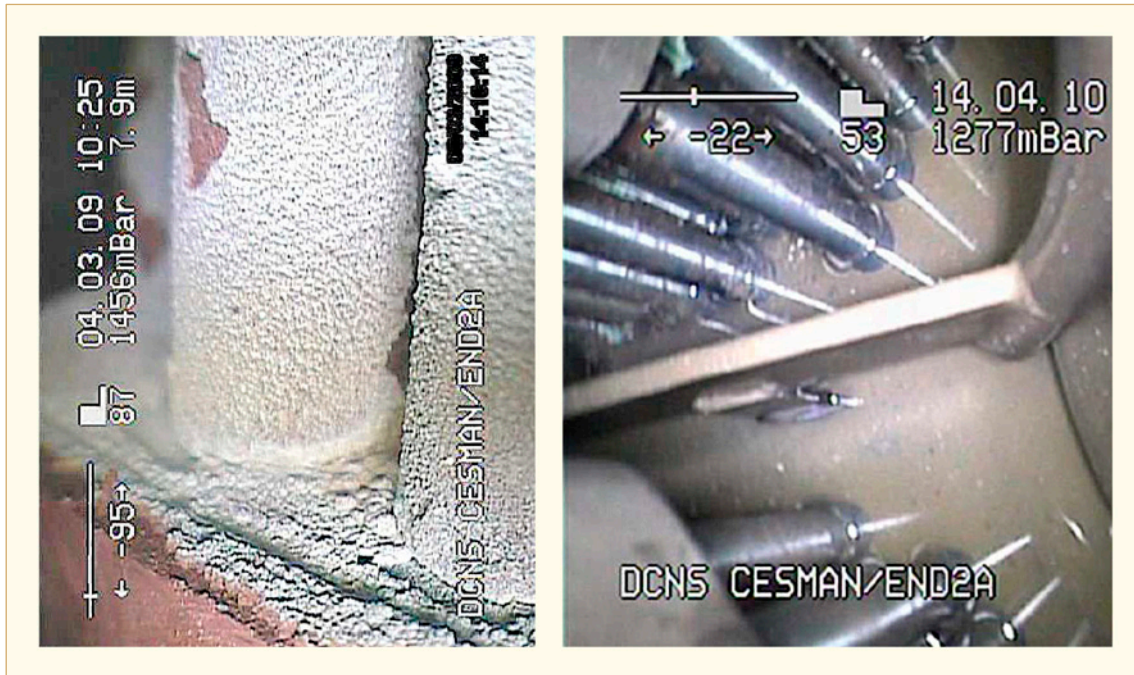


Figure 6-1: Heat exchanger before (left) and after (right) cleaning by foam process [Corrieu et al, 2012].

AREVA also used foam for decontamination of a long metallic pipe (pneumatic transport system) at La Hague, with the great claimed advantage of being able to treat long pipes that other liquid chemicals such as acidic or oxidizing solutions cannot treat; in addition, foam is producing less amount of wastes with a high decontamination factor with a reduction of activity level below 1 Bq/cm<sup>2</sup>. The tests have been done with aluminium using a mixture of phosphonitric acid but may be applied to other metallic surfaces by changing the foam formulation [Damerval et al, 2012].

AREVA also used laser ablation for decontamination with the advantage for this dry process of wastes minimization and avoiding the use of chemicals or abrasives or disks. The principle of the photonic LASER process is absorption of the photon energy by the contaminant. The addition of a vacuum system allows to trap contamination onto filters, thus avoiding any dissemination of contamination in the room where the process is implemented [Moggia et al, 2012]. The principle and example of application are shown on Figure 6-2.

AREVA finally explained the NiThrow™ approach for maintenance activities based on cryogenic process (3500 bars, -140°C).

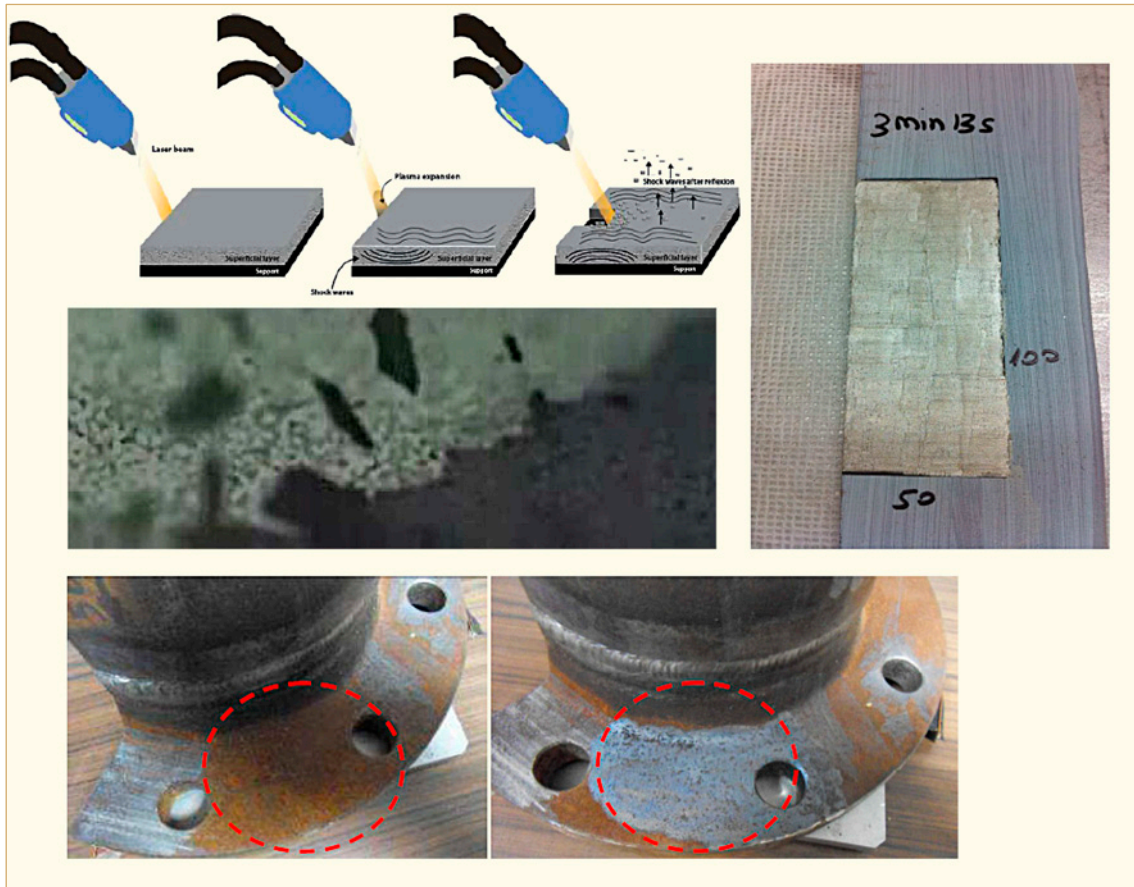


Figure 6-2: AREVA Laser decontamination by, with ablation principle (above top), efficiency on one piece (above right), particle ejection due to laser beam (middle) and ablation of iron oxide (before treatment on bottom left and after treatment on bottom right with complete rust elimination) [Moggia et al, 2012].

Paks VVER in Hungary cooperated with Universities to develop soft decontamination of austenitic steel surfaces of various components, with the optimization of the main parameters, i.e. temperature, concentration of chemicals, flow rates, contact time. It may be applied on surfaces covered by resistant Cr-enriched magnetite or having a hybrid structure of amorphous and crystalline phases. In case of samples, which were not decontaminated earlier, the chromium substituted magnetite crystals take part in the formation of a protective oxide layer due to the low solubility of chromites with the reductive water coolant at high temperature. After decontamination the passivity of stainless steel samples did not decrease [Szabo Nagy et al, 2012]. The structure and morphology of the protective oxide layers formed on the inner surface of SG tubes before and after AP-Citrix decontamination solution is illustrated on Figure 6-3.

## 7 Future trends or new developments (NPC 2012) (Francis Nordmann)

### 7.1 Fast Breeder Reactors (FBR)

Several new NPP units are under construction in Russia including the new FBR -800 in construction at Beloyarsk, which will use MOX fuel. There is also 1 FBR in operation, BN-600 which started operating in 1980. After the experimental reactors BR-5/0 (1958-2002) and BOR-60 (since 1969) the commercial FBR program in Russia started in 1960 with BN-350. Lead-Bismuth eutectic, which has been used in military application, is under development.

### 7.2 New generation of PWR

EPRI has performed assessments of water chemistry control guidance for advanced plant design: AP1000 (Westinghouse), US-EPR (AREVA), APWR (Mitsubishi), APR1400 (Korea) ABWR (Toshiba), ESBWR (GE-Hitachi). These new NPP will use materials resistant to PWSCC and with low cobalt.

UK is defining the chemistry and radiochemistry for the 4 new EPR units to be constructed at Hinkley Point B and Sizewell, 2 units on each site, for compensating the nuclear decrease contribution in electricity production.

Enriched Boric Acid (EBA) for the EPR, The AP1000 will have direct injection of H<sub>2</sub> instead of through a control tank, Electro-de-ionization (EDI) instead of Ion Exchange Resins (IER) for SG blow down purification at 0.6% feed water flow. The AP1000 is not including EBA; the Westinghouse representative claimed that the risk of AOA might be higher if the quantity of precipitated B would be the same but with a higher B-10 enrichment, while several attendees considered that EBA, allowing a lower total B and Li content would, on the opposite, decrease such a risk. In fact, the absence of EBA in AP 100 seems to be explained by the absence of primary water waste recycling

All the design modifications have been evaluated by EPRI to see if there is any impact on chemistry by comparing to existing chemistry guidelines what would be OK, need some changes, not enough or too restrictive, do not work or missing. However, ANT-International made a comment that a bird-eye expert and international view should be taken to look at some deeper improvement or more drastic modification rather than just looking at existing parameters to check what is working or not.

AREVA presented its new water chemistry guidebook for new PWRs which includes EBA, for pH/Li better compatibility with fuel options and  $\text{pH}_T \geq 7.2$  and H<sub>2</sub> range of 17-28 which is sufficient to be above Ni/NiO transition ensuring Ni stability.

EDF briefly explained the design improvements in the EPR of Flamanville 3 under construction, for a 60 years lifetime, with adequate materials to avoid FAC, SG fouling and TSP blockage, start-up purification system, injection system for 2 amines, potential dispersant injection, chemical hazard precaution, etc.

EDF made a presentation explaining the benefit and objectives of using EBA in the EPR FA3 of Flamanville 3 [Cossé et al, 2012]:

- Smaller volume of tanks or lower concentration of boric acid, thus less risk of crystallization;
- Application of an evolutionary chemistry allowing to maintain a constant  $\text{pH}_{300^\circ\text{C}}$  control of 7.2 throughout the entire cycle, as shown on Figure 7-1, with its corresponding advantages described in other documents and Section 2.7.3 (ORE, etc.);
- Reduction in boric acid wastes with a maximum recycling.



The various systems will use a 37% B-10 enrichment but, in case of make-up, for B-10 depletion compensation (B-10 consumption from the reaction  $^{10}\text{B} (n, \alpha) ^7\text{Li}$ ), a higher enrichment will be selected (> 90% in atom), called VEBA, Very Enriched Boric Acid. Other boron losses are from leaks and dilution (boron decrease).

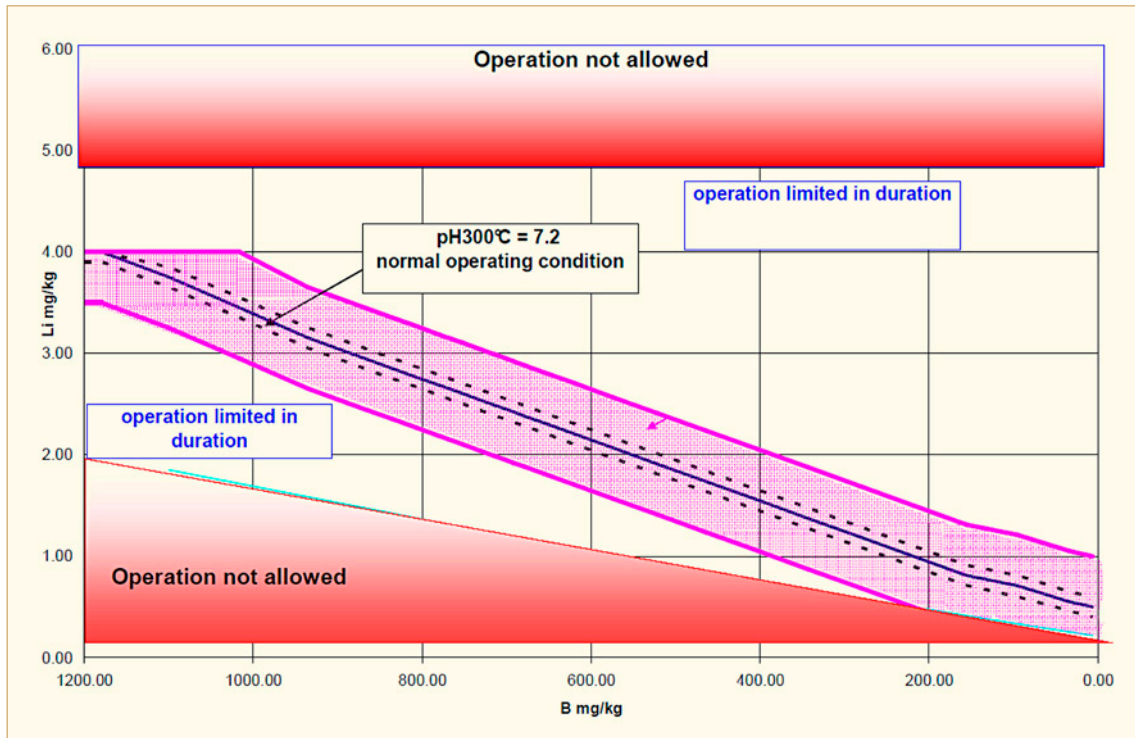


Figure 7-1: Boron / Lithium diagram for EPR of Flamanville 3 [Cossé et al, 2012].

EDF also described the strategies for auxiliary systems of the EPR™ (Flamanville 3) and UK-EPR™. One of the most significant improvements of the design is to avoid the risks associated with the hydrogen and oxygen mixtures thanks to the TEG (gas effluent treatment) system in charge of H<sub>2</sub>/O<sub>2</sub> risk prevention. This includes: (i) flushing with nitrogen, (ii) catalytic recombiner to efficiently reduce H<sub>2</sub> and O<sub>2</sub> concentrations in the flushing gas and (iii) controlling H<sub>2</sub> and O<sub>2</sub> on-line upstream and downstream the recombiner with automatic action initiated if H<sub>2</sub> > 4% or O<sub>2</sub> > 2%.

Two treatments are proposed by AREVA for the secondary system, either H-AVT or amine (morpholine or ETA). ANT-International made a comment that the N<sub>2</sub>H<sub>4</sub>/O<sub>2</sub> ratio ranging from 5 to 8 is too low and should be > 8 and at least 20 ppb N<sub>2</sub>H<sub>4</sub>. The new diagram for Sodium (ppb) and cation conductivity at SB blow down, during normal power operation, shown on Figure 7-2 is taking into account the presence of more resistant materials for SG tubing, and avoiding unnecessary restrictive values [Ryckenlinck et al, 2012].

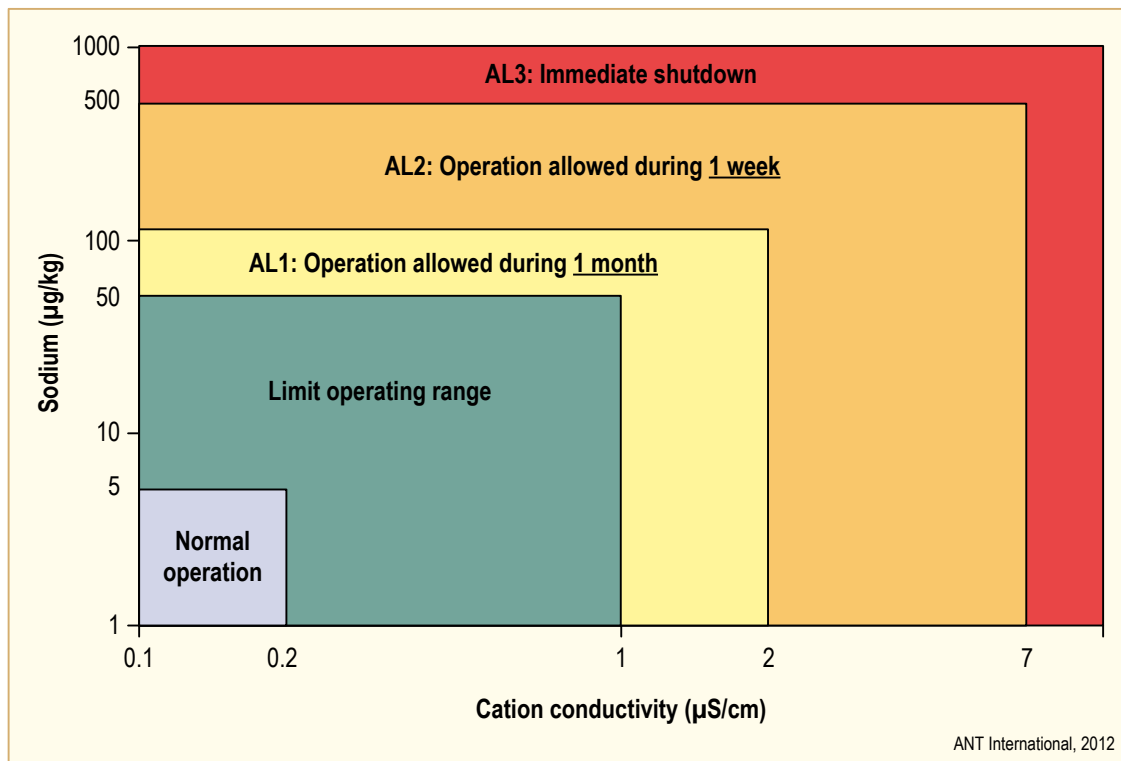


Figure 7-2: Cation conductivity / sodium diagram at SB blow down during for new AREVA PWR [Ryckenlinck et al, 2012].

A Canadian University made calculation and measurements of pH for LiOH solutions and corrosion (metal loss) of stainless steel up to 650°C, for Supercritical water-cooled reactors.

The CRV Laboratory of Rez Research Centre in Czech Republic described the Supercritical Water Loop (SCWL) which is able to test materials, chemistry, radiolysis, etc, at 600°C and 25 MPa.

Bohunice (Slovak Republic) developed a new chemical information system for supporting chemists in early detection and identification of anomalies in the primary and secondary systems of this VVER-440.

### 7.3 New generation of BWR

Hokuriku Electric Co of Japan presented the technology for ORE reduction in Advanced BWR (ABWR), which has no piping for Reactor recirculation system, the largest source term of radiation in conventional BWRs and where carbon steel (CS) piping of the Reactor Water Cleanup System (RWCU) and Residual Heat Removal System (RHRS) are the largest source. The reduction process has been applied in Shika-2 (1 of the 4 ABWR units in Japan) for carbon steel surfaces. Before loading, an alkaline pre-filming was applied to RWCU during plant start-up testing; then after start of operation, FW iron concentration control was applied, both resulting in ORE reduction as expected with 0.35 man.Sv at 3<sup>rd</sup> outage and an equilibrium extrapolated low value of 0.5 man.Sv [Saito et al, 2012].

The alkaline pre-filming is able to decrease oxide build-up by a factor of 1/3 on CS. The reactor water temperature was set between 200 and 230°C with a pH between 8 and 8.5 during 87 hours. Feed water iron concentration and more generally corrosion products concentration plays a role as the trapping site for Ni and Co, which becomes <sup>58</sup>Co and <sup>60</sup>Co. It has been noted that the Ni increase from 1 to 5-6 ppb induced a slight decrease of <sup>60</sup>Co deposited on stainless steel but a large increase on carbon steel as illustrated on Figure 7-3. At the beginning, Co-58 increased at Shika-1 due to insufficient Fe and then decreased when Fe became sufficient.

## 8 EPRI 2012 Steam generator secondary side management conference (Dewey Rochester)

### 8.1 General information

The key presentation was an historical one, with the following objectives:

- 1) To review significant secondary side experiences (including PWSCC caused by denting) to remind and inform SG engineers, operators and chemistry personnel of the occurrence of these events and of the lessons learned.
- 2) To help minimize chances of similar events in the future.

This review acknowledged that the secondary side experiences were incomplete, e.g., it focused most heavily on Westinghouse-type SG designs, including those of licensees, since these are the most common. This list of experiences serves as the start of a “living” list of events that the industry will maintain and expand for educational and training purposes. This presentation contained a historical summary of all the steam generator secondary side problems for the various tube materials that have been in service over the past fifty years. The tubing materials included stainless steel (Shippingport), Alloy 600MA, sensitized Alloy 600MA, Alloy 600TT, and Siemens 800NG [Gorman, 2012].

### 8.2 Deposit generation and transport

#### 8.2.1 Corrosion products during start-ups

An EPRI presentation provided information on the quantities of iron transport to the steam generators (SGs) during plant start-up and lay-up periods. The goals of these efforts are to reduce the SG deposits and sludge build-up in PWRs and corrosion product transport to the BWR fuel. Over the years, various lay-up practices have been employed at nuclear and fossil plants with varying degrees of success. The most recent EPRI document (EPRI 1015657 - 2009) gives three guiding principles:

- 1) Maintain the chemical oxidation-reduction potential of the water in the cycle the same at all plant conditions.
- 2) Keep the water from becoming oxygenated.
- 3) Keep water and moisture out of the secondary components that are to be maintained in a dry condition during the shutdown.

The corrosion rate (mg/dm<sup>2</sup>-month) of carbon steel increased respectively from 0.3 to 0.9; 5.0 and 41 for a relative humidity of 50%, 70%, 90%, and 100%. It was of 400 mg/dm<sup>2</sup>-month when flooded with oxygenated water. Carbon steel corrosion for drained equipment ranged ~40 mg/dm<sup>2</sup>-month to 1 for dehumidified equipment [Fruzzetti, 2012a]. Numerous cycles for BWRs and PWRs were reviewed to estimate the amount of iron transported during start-up, defined as the first two weeks of the cycle. For PWRs, the median iron was reported to be 12% of the total iron transported in the SG during a fuel cycle, equivalent to 9.1 kg of iron oxide, with a high of 20% (52.6kg) and a low of 2% (2.3 kg). For the plants reporting, a median value of iron oxide transported during start-up was 136 kg with a high of 316 kg and a low of 45.8 kg. For BWRs, the amount of iron transported during start-ups was considerably less, as shown on Figure 8-1.

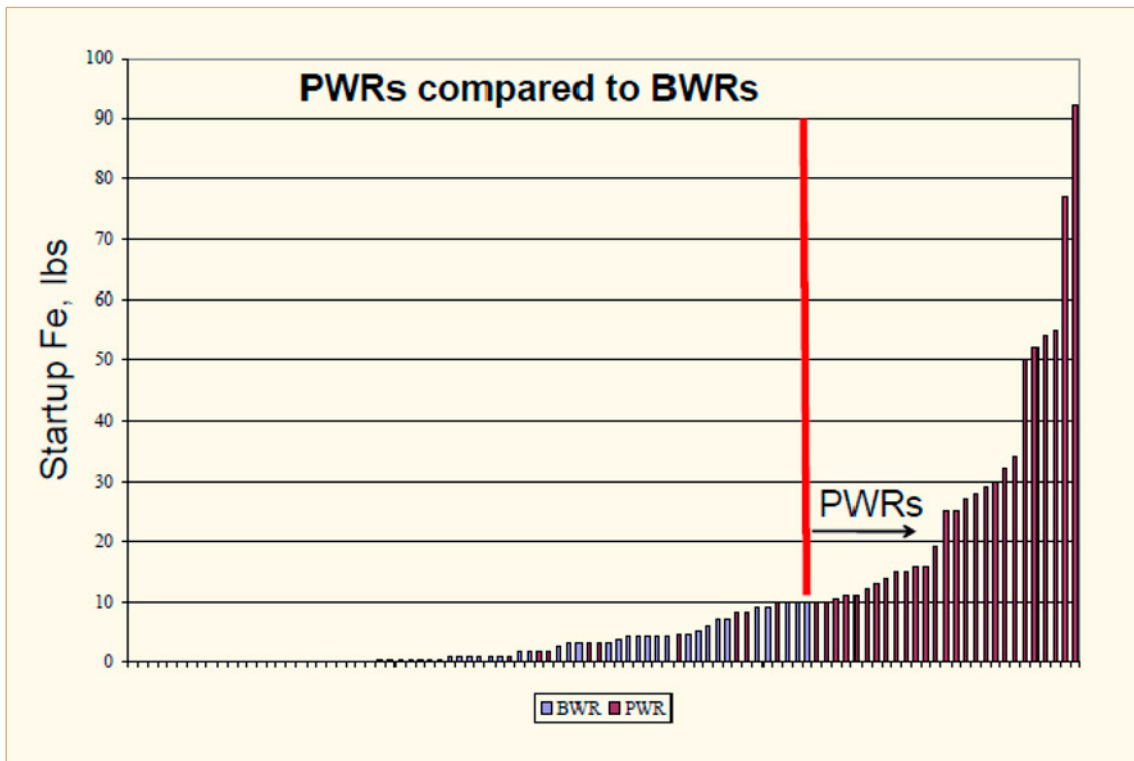


Figure 8-1: Start-up iron comparison between PWRs and BWRs [Fruzzetti, 2012a].

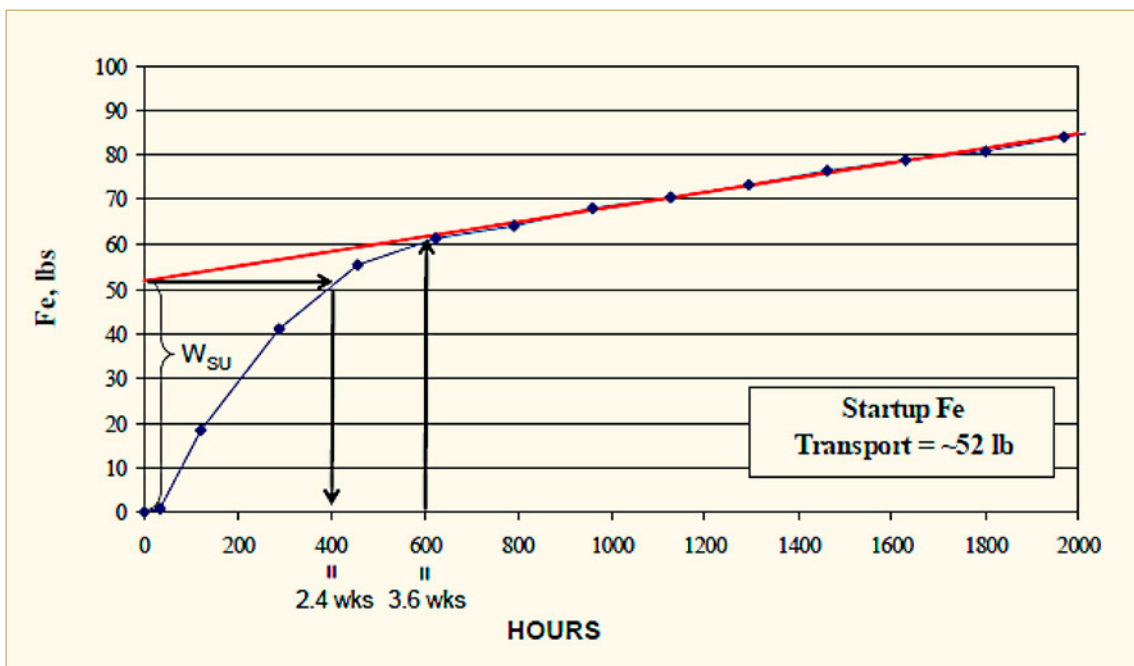


Figure 8-2: Start-up iron calculation at plant A [Fruzzetti, 2012a].

EPRI concluded that the impact of start-up iron transport at PWRs can be significant even when full flow polishers are used. Large variations occurred at plants of similar design with similar lay-up and start-up practices and the major source of iron appears to be from the high pressure heaters but is often plant specific. However the amount of iron transported to the SGs during start-up appears to be relatively small (see Figure 8-2) compared to the total amount of iron transported during the cycle [Fruzzetti, 2012a].

## 8.2.2 Impact of corrosion products on SG corrosion

Bruce Unit A2 is a CANDU designed pressurized heavy water reactor. The unit was shut down in 1995 and was returned to service in 2012 after extensive maintenance, which included:

- Pressure tube and calandria tube replacement – CANDU core structure
- Steam generator replacement with Alloy 800 tubing
- Electrical systems upgrade
- Main condenser refurbishment
- Feed water heater replacement
- Shutdown System 2 (SDS2) enhancement
- Many other significant maintenance items

In February 2012 during pre-start-up activities, high concentrations of copper were present in the balance of plant. Many copper containing components had been replaced during the outage but a significant amount of legacy copper remained. Some copper had already entered the replacement steam generators. A risk assessment was performed to determine the best course of action considering schedule delays, potential life reduction of the new SGs and near, intermediate and long term actions [Little, 2012c].

It was estimated that about 75 gm of soluble and suspended copper would immediately enter the SGs and that potentially 50 kg of copper deposits were present in the condensate/feed water systems. The approach, therefore, was risk minimization versus copper removal.

A literature review found little data on copper corrosion of Alloy 800 although Alloy 800 is less susceptible to copper corrosion than Alloy 600MA. Copper oxide is recognized as a deleterious species in the secondary side but copper metal is not. A comparison of Alloy 800 and Alloy 600MA in oxidizing environments was performed.

The concern is that during operation, outages, start-ups and shutdowns the changing environments from reducing to oxidizing and back will lead to oxidation of copper to copper oxide and the reduction of copper oxide to copper thus causing tube degradation.

Testing by Laborelec concluded:

- Without reducing agents and in the absence of lead, the susceptibility of Alloy 800 is about an order of magnitude less than that of Alloy 600, i.e., the improvement factor is on the order of 10.
- In general, Alloy 800 is much less sensitive to oxidizing conditions than Alloy 600. This is evident from the significant reduction in susceptibility of Alloy 600 when hydrogen is present and the relative absence of an effect from hydrogen for Alloy 800.
- Despite being more resistant than Alloy 600, the presence of copper oxide can reduce the Alloy 800 tube life by at least one third and possibly more.

## 9 Conclusions (Francis Nordmann and Dewey Rochester)

Fukushima accident had a significant impact on political decision for the future of number of operating NPPs. It also had an influence on various studies (less investment), mainly in countries where there is a decision toward decreasing the number of operating units.

The development of new types of reactors is nevertheless active moreover since several countries keep an active program of building new reactors such as EPR, AP1000, ABWR, VVER-TOI.

### 9.1 Primary coolant

The dose rate mitigation or Occupational Radiation Exposure (ORE) remains a key point within studies and improvements with the following remedies that have been covered:

- SG tubing manufacturing is likely the most important way of decreasing further ORE; the surface of SG tubing through the manufacturing process is thus more important than the SG tubing material composition itself, as thought in the past due to the difference in nickel content which is activated into Co-58;
- Passivation during Hot Functional Tests (HFT) potentially followed by a cleaning phase (that has not yet been very successful) is an additional way for decreasing ORE;
- Zinc is added in a growing number of units, mainly for ORE and will be also included in the new type of VVER (VVER-TOI), which has another characteristic of western PWR units, with the direct addition of hydrogen instead of using ammonia.

A very large number of studies have been performed to evaluate the characteristics and composition of crud deposited on the fuel, since this may have a tremendous impact on:

- Potential AOA/CIPS, a subject particularly sensitive with high performances fuel options;
- Influence on radioactive corrosion products, and thus ORE;
- Heat transfer from the fuel cladding and its corrosion risk
- Efficiency of zinc addition on dose rate reduction or corrosion mitigation.

The use of Enriched Boric Acid (EBA) has been shown to be a valuable option, particularly with high performances fuels, which require a high B-10 amount at Beginning of Cycle (BOC). This is the way to operate with the optimum  $\text{pH}_T$  while keeping Li value within the maximum value allowed by fuel vendors or limited for other reasons. EBA is included in the EPR design but not in the AP1000, due to the absence of primary water waste treatment and recovery.

For BWR chemistry, SCC mitigation remains the very high priority with always improved strategies, the last one being the Early Hydrogen Water Chemistry (EHWC) to be in a reducing environment at lower power, when impurities concentration from shutdown is higher. Several other issues are similar to PWR ones (e.g. radiation field and corrosion products control).

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