

Key Emerging Issues and Recent Progress Related to Plant Chemistry/Corrosion in PWR/VVER/CANDU Reactors

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Nomenclature

Unit conversion

1 Introduction

The NPC 2014 conference took place in Sapporo, Japan, 26-31 October 2014. It was the 19th conference in the series of “Nuclear Power Plant Chemistry” that started in Bournemouth in 1977 and is now taking place every other year, alternately in the following loop: Europe, Asia, Europe, and America. The previous one, NPC 2012, was in Paris in September 2010, and the conclusion and key facts were presented at LCC8 seminar in 2013.

The next conference will be in the original country, UK, however, a different city, Brighton 1-6 October 2016.

This report is also briefly covering a one day workshop held in Paris, on November 21, 2013 by SFEN, on how the environment is taken into account by chemistry of French NPP.

The NPC 2014 conference report is divided in two parts:

- One is related to PWR, VVER CANDU, HPWR reactors as well as some of the various other topics more or less independent from the type of reactors, such as maintenance, fuel, future trends, auxiliary systems, wastes, etc. It is covering 122 papers, and thus helps the reader at having a summary of the key facts of this conference that may be of use of Utilities, R&D Laboratories, vendors, Regulators. The information is gathered by technical subjects with additional point of view of the author when appropriate, rather than listed one after the other, which would be of no benefit.
- Another report prepared by S. Hettiarachchi is covering BWR, Fukushima event and its treatment, as well as a various other topics more or less independent from the type of reactors, such as maintenance, fuel, future trends, auxiliary systems, wastes, etc. This other report covers 105 papers.

There have been 12 oral sessions in addition to special sessions for Fukushima, covering respectively 68 and 24 papers, in addition to poster sessions dealing with 152 papers including 22 for Fukushima. However, 9 posters and 8 oral presentations were not associated with a paper distributed in the conference CD-ROM proceedings and, consequently, are not covered here. The overall number of papers is thus 244 with 227 associated with a written paper in the CD-ROM.

After the opening speeches, there have been three keynote lectures from the three main historical nuclear countries, i.e. USA, France and Japan. They are giving an interesting overview of the important facts in each country.

Japan, the hosting country, explained the organisation of chemistry, the various involved committees and overall activities including members from Industry (Plant Utilities), Academia and Universities [Katsumura, 2014]. Standards and guidelines have been developed by the Standard Committee of AESJ in accordance with the Standard Committee Rules, which assure fairness, impartiality and transparency in the process of deliberating on standards. The Water Chemistry Division edited the first “Water Chemistry Road Map” in 2007 and revised the second one in 2009, as shown on Figure 1-1. The fuel behaviour is of high importance in it, while radiolysis, a domain where Japan has always been very active, being at the initiative of the international radiolysis workshops after each NCP conference, is also included. The new one has been delayed and will take into account the lessons learned from the Fukushima Nuclear Accident.

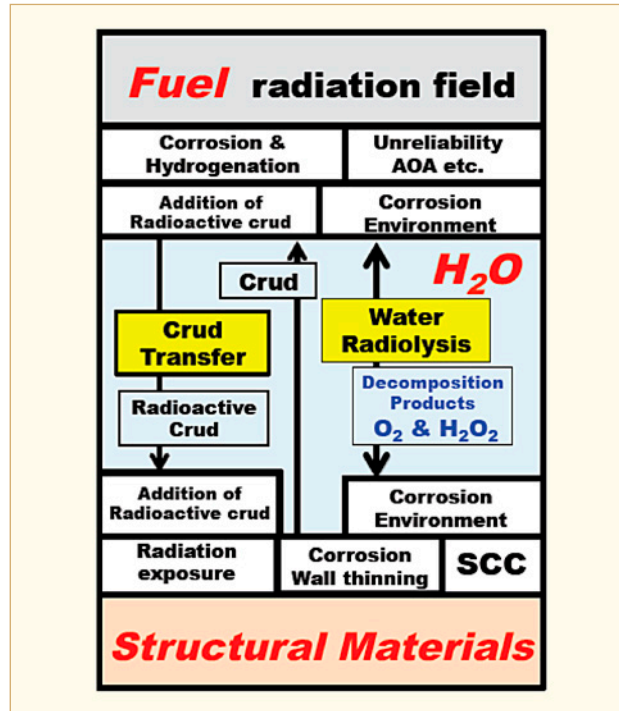


Figure 1-1: Schematic relation between fuel, structural material and coolant water through water radiolysis and crud transfer [Katsumura, 2014].

In the USA, EPRI is playing a large role and is giving an overview of what happens in US NPP [Fruzzetti et al, 2014a]. The major trends are operating with a sufficiently high pH, both in the primary coolant for lower dose rates and in the secondary system for mitigating Flow Accelerated Corrosion (FAC) and corrosion transport and deposition in the steam generators (SG). In the primary coolant, zinc addition is an additional way to minimize dose rates, while in the secondary system; addition of dispersants may also contribute to minimize corrosion products deposition in the SG.

The general trend on chemistry at EDF has been explained at the November 2013 SFEN workshop. The main issues of primary coolant at EDF NPP are contamination and dose rates (ORE), corrosion product transport, AOA due to deposit and material integrity while the solutions are reaching the target pH at the very beginning of the fuel cycle, optimum Li and H₂ concentrations, optimized shutdown process, zinc addition, SG tubing passivation at start up. The main issues for the secondary system are FAC of carbon steel, IGA/SCC of Alloy 600 SG tubes, oxides deposition in SG, while the solutions are an optimum pH, amine selections, and optimum N₂H₄ concentration of this hazardous chemical and preventive cleaning of SGs. On the condenser cooling system, the main issues are fouling and power decrease, cooling capability while the solutions are acid injections or polyacrylate (for fouling), dispersant (for suspended solids), biocide addition or electro chlorination (for amoebae and legionella) [Bretelle, 2013].

2 Primary water chemistry

2.1 Guidelines and chemistry optimisation

The PWR primary chemistry in the USA focuses at increasing pH_T (necessitating increased lithium concentrations) as well as a shift to more constant pH_T programs (as compared to modified pH_T programs in which the lithium concentration is maintained at a constant value for any portion of the cycle, thereby allowing the pH_T to increase as the boron concentration decreases). There are now about half of US PWR operating at elevated pH (>7.2), as illustrated on Figure 2-1.

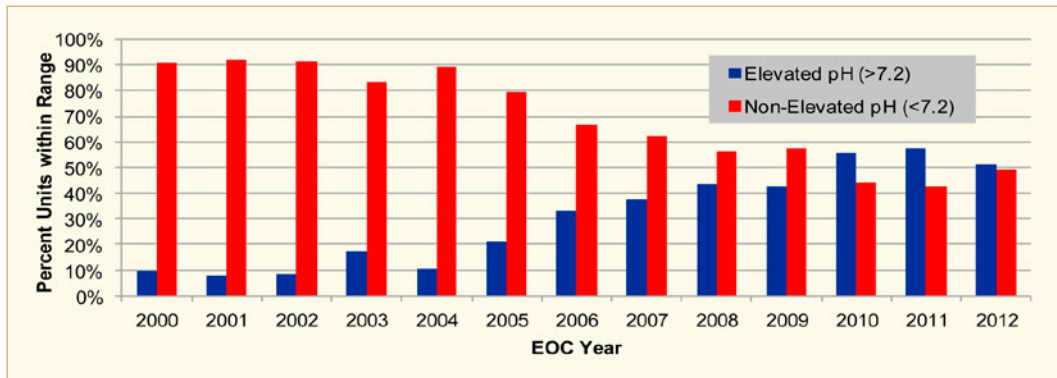


Figure 2-1: Evolution of PWR primary pH_T (elevated vs. non-elevated) in the U.S. [Fruzzetti et al, 2014a].

EPRI produced Revision 7 of the EPRI PWR Primary Water Chemistry Guidelines in order to update the document to ensure it contains the state-of-the-art of technical information and industry operating experience.

An Institute in charge of fuel in Ukraine studied the possible improvements in primary water chemistry of VVER, examining the practices and new trends in PWRs [Krasnorutskyy et al, 2014].

The most promising approach is the replacement of ammonium injection by introduction of gaseous hydrogen into the primary coolant. This will reduce expenses for ammonium and hydrazine as well as quantity of radioactive waste. The most important factor, however, is that hydrogen injection will allow a better responsiveness for primary coolant chemistry management during unit transients and significantly reduce the load on the purification system, SVO-2.

The quantity of consumed boric acid and releases into the environment is considerably higher in Zaporozhie-3 (Ukraine), with 17000kg/year, than in PWR units, with for example 120 kg/year in Emsland, a German Konvoi operated with EBA at 28% B-10 enrichment. Although EBA has been considered of very high importance in Ukraine, with new fuel performances and MOX possibility, its use cannot be decided due to the cost which would be unacceptable without a drastic recycling of boric acid and after completing the entire set of measures to upgrade the primary coolant chemistry.

Replacement of the KOH by LiOH is considered to have a positive effect on radioactive waste amount reduction. Such replacement will help to reduce the amounts of radioactive waste by 1.3 tons a year. However, the use of LiOH with natural Li would increase emissions of tritium into the environment, while enriched Li-7 would increase the expenses of products to be bought from foreign manufacturers. Then, it should be considered that such a replacement may increase the corrosion rate of E110 fuel rod cladding of VVER, although it would remain acceptable.

Zinc injection does not adversely affect zirconium alloy corrosion of fuel cladding and helps at reducing stainless steel corrosion rates, as shown on Figure 2-2. Injection of zinc in VVER may reduce the activity of primary circuit inner surfaces due to lower amounts of corrosion products. Zinc, however, may displace cobalt from oxides accumulated over a long-term operating period in high-temperature titanium filters of the SVO-1 water treatment system, which would increase coolant activity. Nevertheless, with the activity increasing with time, Ukraine is considering implementing countermeasures. As for EBA, without preliminary measures to upgrade the primary coolant chemistry and recycling, the consumption of zinc acetate would be high as compared to PWR, again resulting in expenses of products to be bought from foreign manufacturers. Finally, in VVER reactors, Co activity accounts for <50% of the total activity. Therefore, zinc injection will not produce an equally tangible and quick effect as in PWRs [Krasnorutskyy et al, 2014].

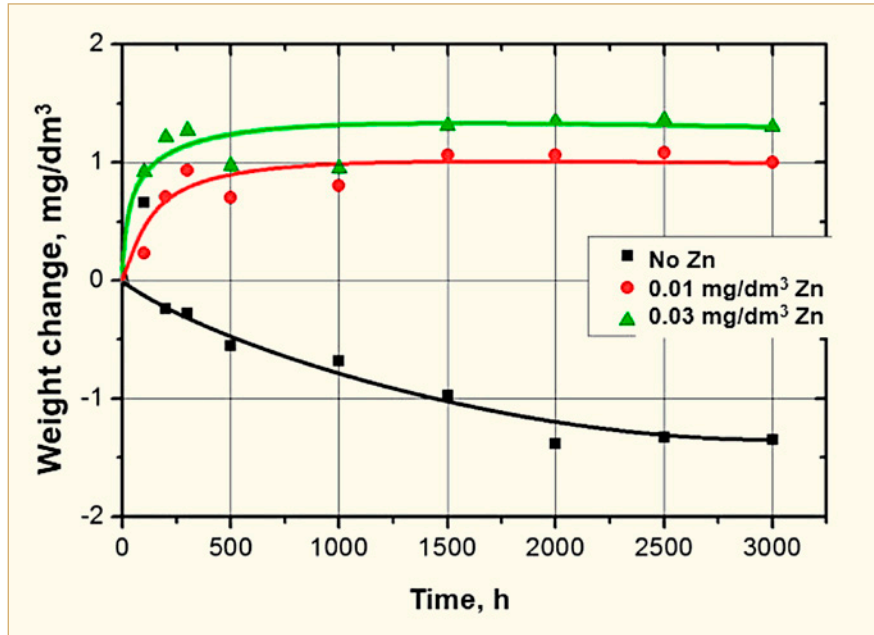


Figure 2-2: Corrosion kinetics of stainless steel Ch18Ni10Ti (18% Cr, 10% Ni, stabilized with Ti) in WWER-1000 primary coolant model medium without Zn and with 0,01 and 0,03 mg/dm³ Zn during oxidation at 350°C [Krasnorutskyy et al, 2014].

Japanese members of the Atomic Energy Society of Japan (AESJ) decided to establish guidelines for water chemistry for ensuring the reliability of PWR operation and maintenance with improved material and fuel reliability as well as to reduce dose rates [Kawamura et al, 2014]. For control parameters, these guidelines include action levels: 1 (1 week), 2 (24 hours) and 3 (immediate power reduction). This is shown on Table 2-1 for the primary coolant during power operation. Make up water diagnosis parameters and recommended values are shown on Table 2-2, while the paper includes other situations (start-up, shutdown).

KEY EMERGING ISSUES AND RECENT PROGRESS RELATED TO PLANT CHEMISTRY/CORROSION IN
PWR/VVER/CANDU REACTORS

Table 2-1: Control, conditioning and diagnostic parameters for primary coolant during power operation, after [Kawamura et al, 2014].

Parameters	Control item	Conditioning	Diagnostic	Action levels		Conditioning values	Recommended values	Measurement frequency ^{a)}
				Level	Value			
Conductivity at 25°C (mS/m)	O			1	Setting value ±0.5		-	D
				2	>4			
				3	-			
pH at 25°C	O			1	Setting value ±0.5		7.2-7.4 (at 285°C)	D
				2	<4, >11			
				3	-			
Chloride (µg/L)	O			1	>50		≤10 (or low value at each unit)	W
				2	>150			
				3	>1500			
Fluoride (µg/L)	O			1	>50		≤10 (or low value at each unit)	W
				2	>150			
				3	>1500			
Sulphate (µg/L)	O			1	>50		≤10 (or low value at each unit)	W
				2	>150			
				3	>1500			
Lithium (mg/L)	O	x		1	<0.2, >3.5		-	W
				2	>6			
				3	-			
Boron (mg/L)		x		1		= f (neutronics design and reactivity)		W
				2-3				
Dissolved hydrogen (cm ³ /kg)	O	x		1	<25		-	W
				2	<15, >50			
				3	<5			
Dissolved oxygen (µg/L)	O			1	>5		-	M
				2	>100			
				3	>1000			
Silica (mg/L)			x	1			≤0.5	M
				2-3				
Ammonia (mg/L)			x	1			-	M
				2-3				
γ isotopic (Bq/cm ³)			x	1			Depend on unit specific	M
				2-3				
Iodine-131 (Bq/cm ³)			x	1			-	W
				2-3				
Xenon (Bq/cm ³)			x	1			-	0.5 M
				2-3				
Tritium (Bq/cm ³)			x	1			Depend on unit specific	M
				2-3				
Metallic impurity (µg/L)			x	1			Depend on unit specific	M
				2-3				
Turbidity (mg/L)			x	1			-	3 M
				2-3				

Notes: ^{a)} D: 1 time/day, W: 1 time/week, 0.5 M: 2 times/month, M: 1 time/month, 3 M: 1 time/3 months

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In addition to these guidelines, two main aspects have been selected for Long-term Strategy [Kawamura et al, 2014]. These are:

- **Zinc Injection.** It has been applied to 21 PWRs in Japan to reduce radiation field build-up. Since no adverse effects of zinc injection on Primary Water Stress Corrosion Cracking (PWSCC) for Ni-based alloys have been observed in the field, Zn injection will also be applied in some other units once they will restart [Kawamura et al, 2014].
- **Hydrogen.** An optimized dissolved hydrogen (DH) control in Japanese PWR primary coolant systems has been proposed to reduce radiation field build-up and to mitigate PWSCC initiation and propagation in Ni-based alloys. Several years ago, Japanese PWR utilities considered to apply low DH control for mitigating PWSCC initiation, and examined its effect on IASCC, radiolysis, and fuel performance. Various types of laboratory tests confirmed that low DH (approximately 5 cc-STP/kg-H₂O) was sufficient to suppress water radiolysis and that no adverse effect on the fuel crud deposition and PWSCC of Alloy 600 were observed. Although the test results support the use of a lower DH control, the guidelines still recommend concentrations of 25 to 35 cm³/kg based on reliable experience [Kawamura et al, 2014].

Table 2-2: Diagnostic parameters for makeup water, after [Kawamura et al, 2014].

Diagnostic parameters	Recommended values	Frequency of measurement ^{a)}
Conductivity at 25°C (mS/m)	≤0.2	M
Chloride (µg/L)	≤150	M
Fluoride (µg/L)	≤150	M
Sulphate (µg/L)	≤150	M
Dissolved oxygen (µg/L)	≤300	Q
Silica (mg/L)		Q
Notes: ^{a)} M:1 time/week (or month ?), Q: 4 times/year		
ANT International, 2014		

DIN German Institute for Standardization (DIN e. V.) has a long history of providing industrial standards. Two standards concerning cleanliness in nuclear power plants have recently been revised by a national subcommittee, namely DIN 25410 “Nuclear facilities - Surface cleanliness of components” and DIN 25493 “Nuclear facilities - Protection of metallic surfaces of structural parts from damage from assembly aids, gaskets, packing, packaging material and thermal insulating materials”. The task of the subcommittee was on the one hand to produce documents with sufficient limits for minimization of corrosion, while on the other hand the specified limits had to be technically and organizationally feasible. As a result, both standards reflect the current state of knowledge and technology regarding cleanliness and serve as a framework for good practice. DIN 25493 was subjected to a major revision. Its decision tree, represented on Figure 2-3, defines 4 classes with different limit values which are not based on the product type, but on the application area and application duration [Drexler et al, 2014a].

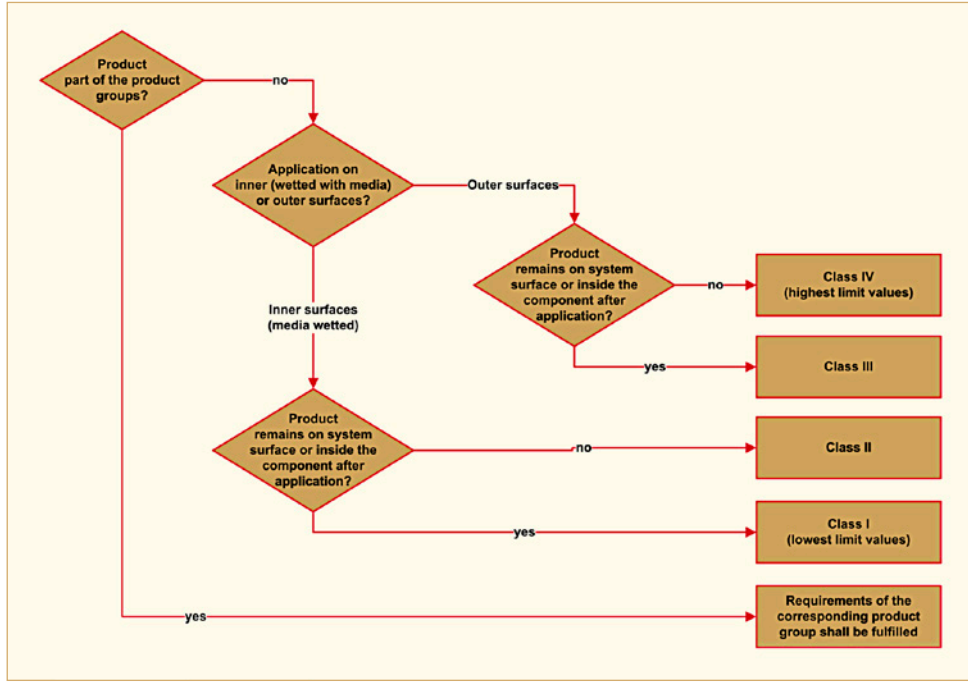


Figure 2-3: Simplified decision tree according to E DIN 25493 [Drexler et al, 2014a].

2.1.1 Hydrogen

The EPRI Guidelines recommendation is to operate in the upper part of the 25–50 cc H₂ (STP)/kg H₂O range for reducing crack growth rates in Alloy 600 and associated weld metals, Alloys 82 and 182. This is not a worldwide decision, Japan being looking more at operating in the lower range of hydrogen, considered as detrimental by EPRI for Primary Water Stress Corrosion Cracking (PWSSCC) of nickel base alloys [Fruzzetti et al, 2014a]. Many countries are keeping the historical value in the middle range of the specification, and practically most of US plants are still operating near the middle of the historical range, Figure 2-4.

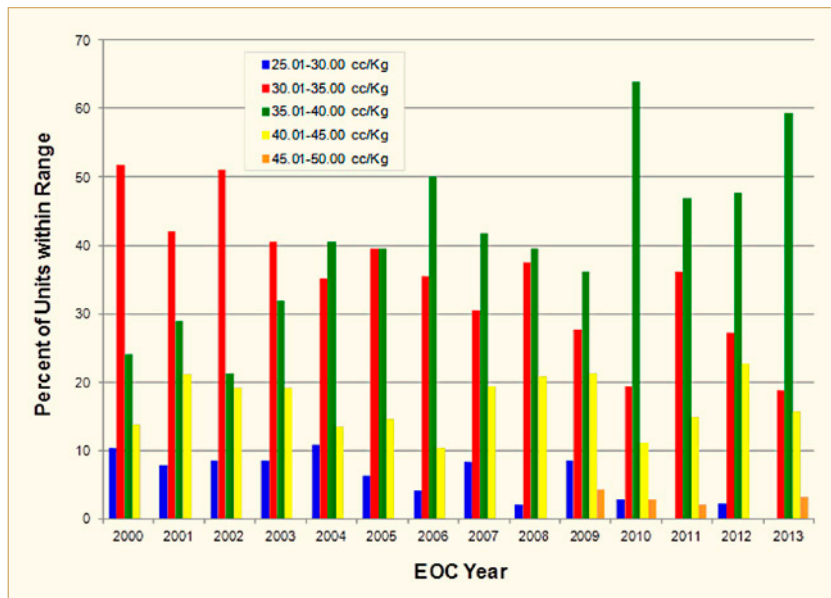


Figure 2-4: Evolution of PWR primary hydrogen in the U.S. [Fruzzetti et al, 2014a].

Besides the various studies on the influence of Dissolved Hydrogen (DH) on SCC of Alloy 600, the Japanese investigated the potential impact of DH concentration on dose rates, through its influence on metallic nickel and nickel oxide on Zircaloy-4. Under lower DH, it is considered that Nickel oxide is deposited mainly on the cladding. The Ni/NiO is at equilibrium for DH concentration considered to be between 15 and 30 cc STP/kg-H₂O at 350°C. The deposition of metallic-Nickel is considered to be suppressed for DH <15 cc, and should contribute to decreasing the radiation source [Nishimura et al, 2014].

Deposition tests on the ZIRLO fuel cladding surface were conducted in Korea to investigate the effect of the concentration ratios of Ni/Fe ions in the primary coolant and thus evaluate the risk of AOA. Ni and Fe ions were injected in a test loop as Ni-EDTA and Fe-EDTA with Ni-Fe concentrations of 20-20ppm, 39-1ppm and 1-39ppm, which means Ni/Fe ratios of 1, 39 and 0.025 at 325°C during 14 days [Baek et al, 2014].

The deposits formed on the ZIRLO surface after the deposition tests were removed and the Fe and Ni concentration were measured by inductively coupled plasma-atomic emission spectrometry (ICP-AES), and then converted to the amount of oxide deposits per unit surface area. The morphologies and chemical compositions of the oxide deposits were analysed using scanning electron microscopy, energy dispersive X-ray spectroscopy and transmission electron microscopy.

For Ni/Fe ratio of 1 (20/20ppm), nickel ferrites with a polyhedral shape were formed. Nickel oxide deposits with a needle shape were formed at high Ni/Fe ratio (39/1), while polyhedral iron oxides with sparse needle-like nickel oxides were formed at low Ni/Fe ratio (1/39). The amount of deposits increased, when Fe-rich oxides were formed, as can be seen on Figure 2-5. The deposition rate of Fe ions was also higher than that of Ni ions. This indicates that deposition of Ni ions is stimulated on the surface of pre-existing Fe-rich oxides [Baek et al, 2014].

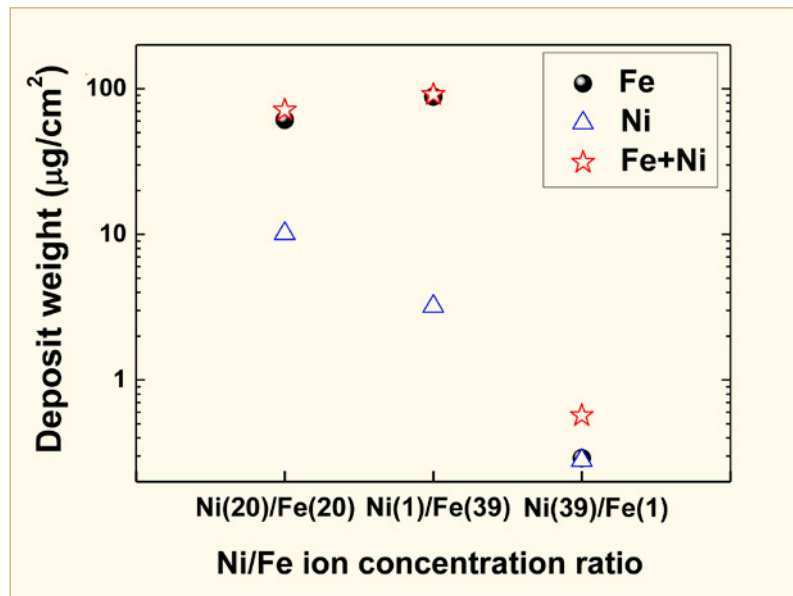


Figure 2-5: Amount of deposits at different Ni/Fe ion ratio [Baek et al, 2014].

Ukraine is considering the use of gaseous hydrogen injection instead of ammonia, hydrazine injection, as a key priority for optimization of primary water chemistry in VVER units (see above Section 2.1).

An optimized dissolved hydrogen (DH) control in Japanese PWR primary coolant systems has been proposed to reduce radiation field build-up and to mitigate PWSCC initiation and propagation in Ni-based alloys. Although test results on radiolysis support the use of a lower DH control, the guidelines still recommend concentrations of 25 to 35 cc/kg based on reliable experience, as explained in above Section 2.1 [Kawamura et al, 2014].

Research in a Korean Institute looked at dissolved hydrogen (DH) influence on corrosion rate of Alloy 690. The purpose was not, as for Alloy 600, to mitigate SCC but more to decrease the release of oxides. The tests at 330°C in various dissolved hydrogen (DH) content (35 to 100 cc/kg) of Li/B solutions and corrosion rates measured by chemical descaling of the oxides [Lee et al, 2014].

As the hydrogen content increased from 35 cc/kg to 100 cc/kg, the corrosion rates of Alloy 690 decreased, Figure 2-6. Field-emission scanning electron microscope (FE-SEM) analyses of the oxide films showed that the sizes of the particles built up on the outer layer decreased as the hydrogen content increased. The difference in particle sizes may be due to changes in the crystal growth environment. The morphology of the oxide films formed on Alloy 690 was of two layers. The compact internal layer composed of very small grains of a few nm in diameter; the external oxide layer is composed of large particles clustered together with varying sizes, from a few tens of nanometres to two hundred nanometres. For hydrogen content of 100 cc/kg, grains seem to diminish from the outmost surface, Figure 2-7.

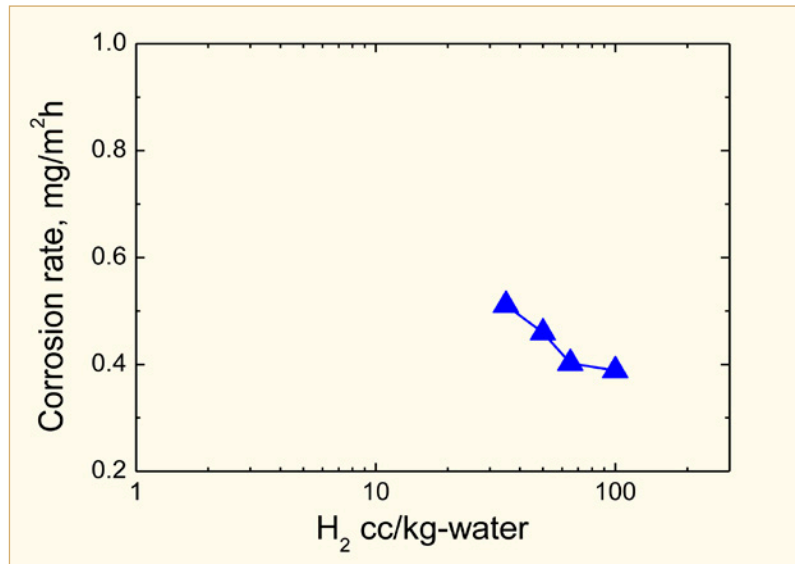


Figure 2-6: The effect of DH concentration on the corrosion rate of Alloy 690 [Lee et al, 2014].

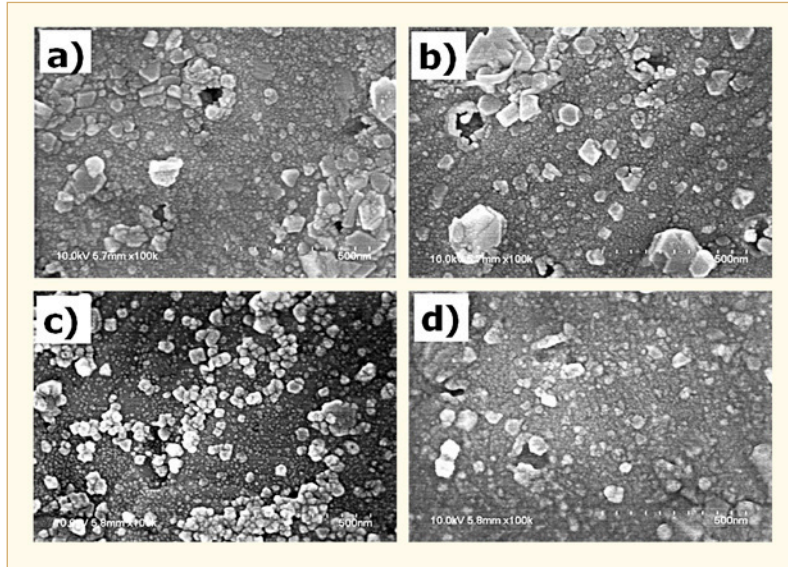


Figure 2-7: SEM morphologies of the oxide films formed on Alloy 690 immersed in different hydrogen concentrations: (a) 35 cc/kg, (b) 50 cc/kg, (c) 65 cc/kg, (d) 100 cc/kg [Lee et al, 2014].

From the X-ray photoelectron spectroscopy (XPS) analyses, it was found that the composition of the oxide films depended on the hydrogen concentrations. For high concentrations of hydrogen, the oxide film exhibited higher chromium and iron content, which were related to the potential zone in which the chromium and iron oxides were stable at testing temperature and pH. At lower hydrogen concentrations, the oxide film showed high nickel concentration, which was related to the Ni/NiO equilibrium potential zone, in which the nickel oxides are stable [Lee et al, 2014].

The Russian designer studied Critical Hydrogen Concentration (CHC) and supposed that neutronic field fluctuations on changing of the radiation-chemical system from the state with low concentration of oxidizing particles over to the state with their high concentrations, may be one of the causes explaining too low calculated value of CHC, as compared with that measured in the experiments in reactor conditions. The correct calculation of CHC in the primary coolant of VVER/PWR should take into account the non-uniformity of n, γ -field in the core, which leads to higher calculated values of CHC. It can be seen in Figure 2-8 that when modelling radiolysis without a short-term neutron irradiation impulse, the hydrogen value of 0.014 mg/kg (0.15 ncm³/kg) is sufficient for suppression of radiolysis while, with the neutron irradiation, even 0.018 mg/kg (0.2 ncm³/kg) is not sufficient [Arkhipov & Kabakchi, 2014].

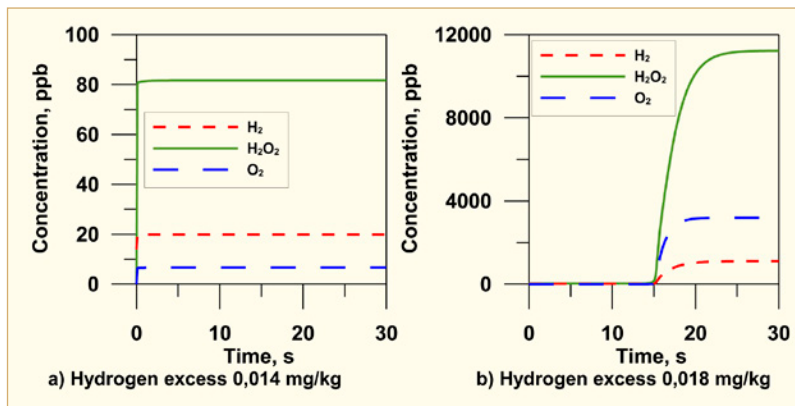


Figure 2-8: Calculation of critical hydrogen concentration (CHC) a) without regard and b) with regard for the disturbing effect of fast neutron impulse [Arkhipov & Kabakchi, 2014].

A Korean Institute looked at the potential influence of dissolved hydrogen concentration on the fuel cladding integrity, through the phase of the zirconium oxide layer formed at the water-zirconium interface. The Raman spectra showed that, in the early stage of the oxidation process, zirconium oxide with both tetragonal and monoclinic phases is formed. With an increase in the oxidation time to 120 h, the unstable tetragonal phase disappears, and the monoclinic phase is dominant. The DH concentration does not have a significant effect on the phase of the zirconium oxide, because the oxide layer formed is a stable one and it acts as a protective layer, retarding further oxidation [Kim et al, 2014a].

Another paper reported tests performed to see if hydrogen concentration, which could be increased for PWSCC mitigation, may have any influence on hydrogen pick up on various fuel cladding materials and spacer weld specimens. M5®, ZIRLO®, optimized ZIRLO™, low Tin ZIRLO™, optimized low Sn Zircaloy-4, and Zircaloy-2 in either as-received or preoxidized surface conditions were exposed for a total of 730 days in high pressure, out-of-reactor autoclave systems operating at three different dissolved hydrogen concentrations, 30, 100, and 500 cc H₂/kg H₂O [Wells et al, 2014].

The tests allowed to conclude that in the absence of a radiation field and at the higher dissolved hydrogen overpressure up to 500 cc H₂/kg H₂O, there was no detectable increase of hydrogen pickup fraction of the Zr-alloys examined, Figure 2-9 and Figure 2-10. Moreover, for each Zr-alloy exposed in the long term experiment, the oxide growth was similar at different DH suggesting that the hydrogen overpressure has no detectable impact on some rate-limiting mass transport process involved among various mass transport processes. Finally, Hydrogen pickup rates for various fuel cladding materials were approximately constant (~50ppm/year-150ppm/year). No correlation between hydrogen pickup rate and DH could be identified for the fuel cladding or spacer weld specimens [Wells et al, 2014].

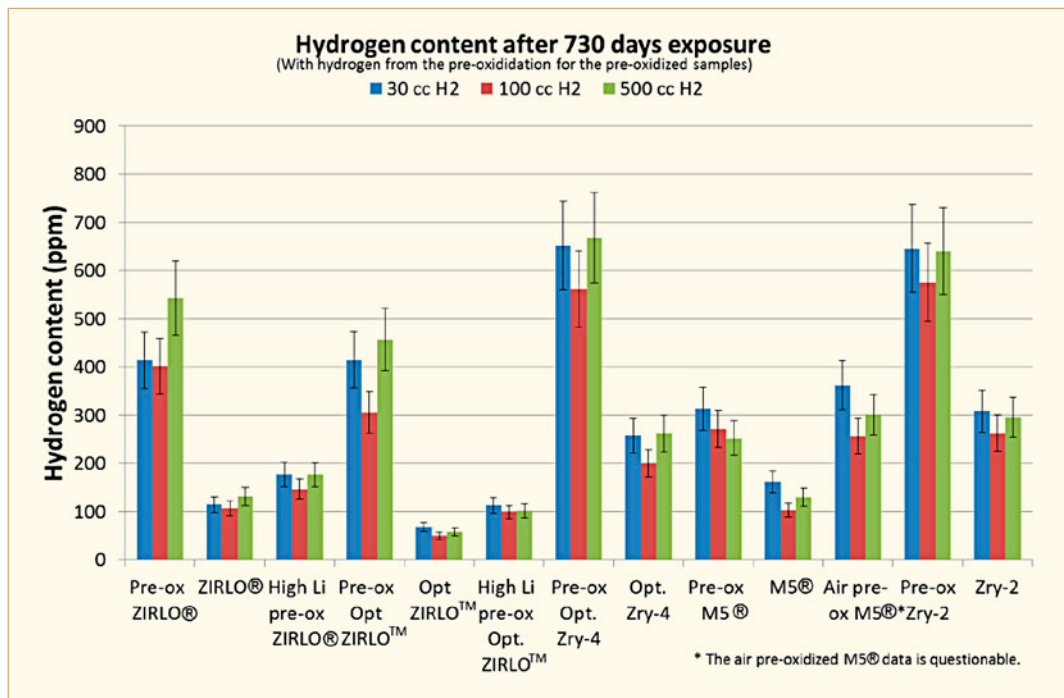


Figure 2-9: Total (including hydrogen from both autoclave exposure and pre-oxidation for the pre-oxidized specimens) hydrogen content in the fuel cladding specimen after 730 days of exposure [Wells et al, 2014].

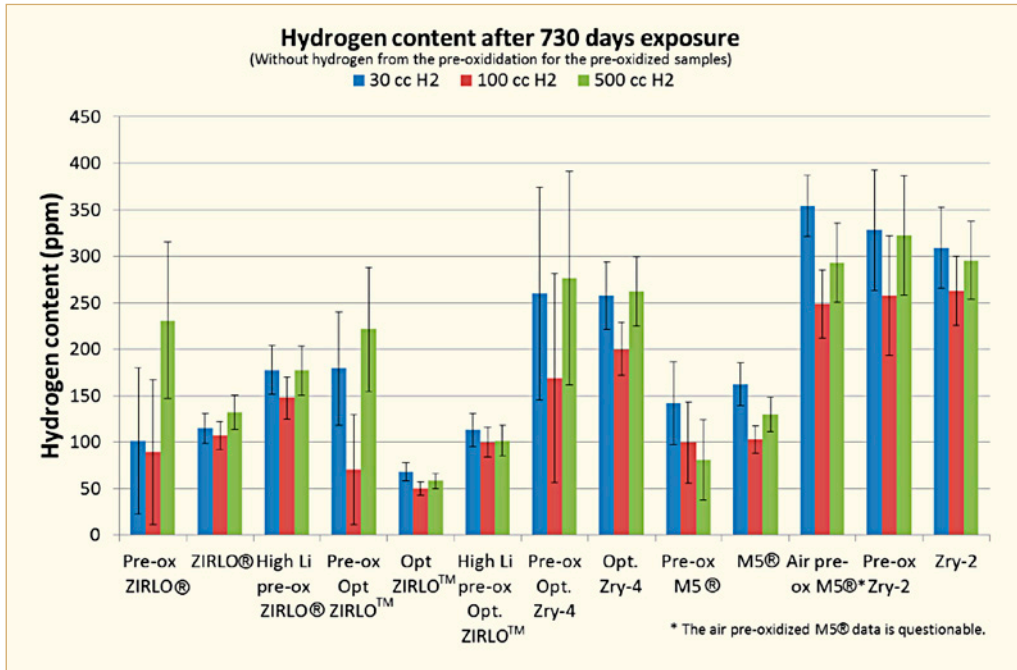


Figure 2-10: Background corrected (for the pre-oxidized specimens) hydrogen content in the fuel cladding specimen after 730 days of exposure [Wells et al, 2014].

Hydrogen (Deuterium) is released in the PHT system storage tank cover gas during full PHT system chemical decontamination of PHWRs. At present, D₂ released in the cover gas is purged out thereby losing the precious D₂O along with tritium. Similarly, sometimes in the moderator cover gas, the D₂ is in excess of the stoichiometric equivalent to oxygen. Consequently, it does not get converted to D₂O in the recombined unit and hence, the concentration of D₂ builds up in the cover gas requiring purging and loss of heavy water and Helium. These losses can be avoided by the use of fuel cell to convert the D₂ formed into D₂O [Chandramohan et al, 2014a].

In a fuel cell, the hydrogen and oxygen are passed through cathodic and anodic compartments and hence direct mixing is avoided and the energy is released in the form of electrochemical energy. For removal of excess H₂ released during decontamination, 4, 6 and 8% hydrogen removal in fuel cell were attempted. The process parameter like the gas flow rate through fuel cell was optimized for gaining maximum recovery efficiency. Feasibility studies for deuterium recovery and further scaled up experiments proved the usefulness of the process [Chandramohan et al, 2014a].

2.1.2 Lithium

Ukraine is considering the use of lithium instead of potassium, but the advantages are not obvious since the VVER fuel cladding corrosion, although acceptable with LiOH, is higher than with KOH. Then, the cost of Li (enriched in Li-7) is far from being negligible while the use of natural Li would induce high tritium releases (see above Section 2.1).

Like in most countries, pH and consequently Li concentration have increased in 11 PWR Japanese units of KEPCO Company, as can be seen on Figure 2-11 [Terachi et al, 2014].

3 Secondary water chemistry

3.1 Guidelines and chemistry optimisation

PWR secondary chemistry in the USA, as in other countries, is focused on reducing materials degradation issues. One of the key points is the optimization of pH to reduce flow accelerated corrosion and thereby minimize corrosion product transport to the steam generators (SGs). Figure 3-1 shows the large application of amines for secondary system treatment in US PWR and a median pH_{25°C} value of 9.75 allowing a median iron concentration of 1.26 µg/kg (ppb) in feed water [Fruzzetti et al, 2014a]. Figure 3-2 demonstrates the benefit of a high pH for decreasing iron concentration, and thus corrosion products deposition in the SG. In addition, dispersant application in the secondary system for better eliminating iron by SG blow down is also a key solution considered by EPRI.

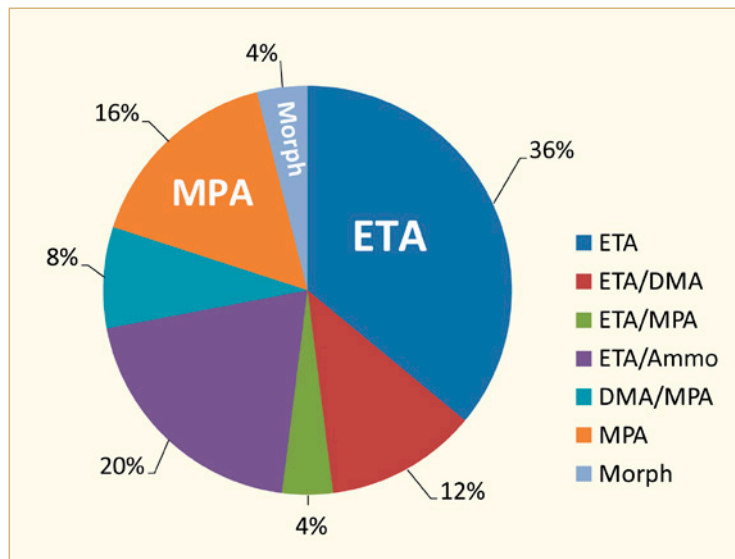


Figure 3-1: Amine use in the U.S. based on EPRI's PWR CMA database [Fruzzetti et al, 2014a].

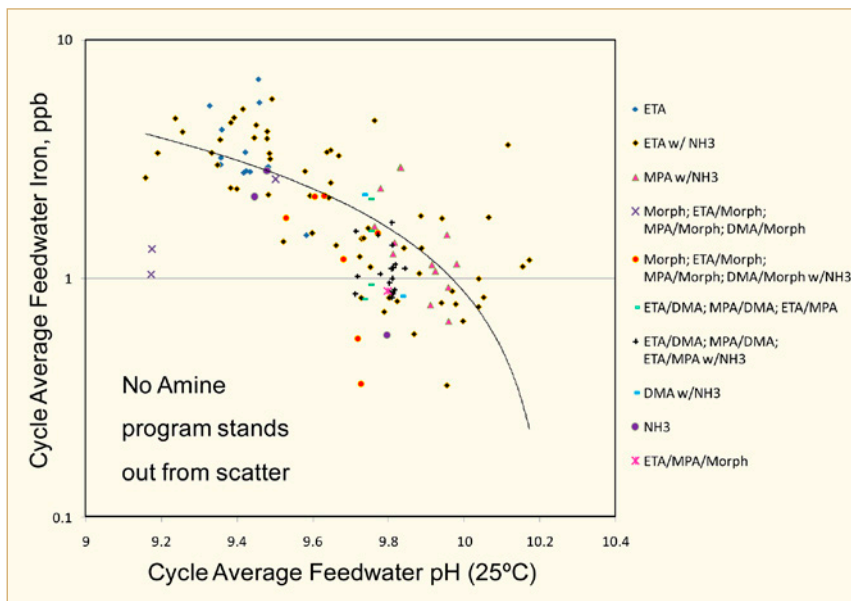


Figure 3-2: Feed water Iron versus feed water pH based on EPRI's PWR CMA database [Fruzzetti et al, 2014a].

After refurbishment of Point Lepreau CANDU Plant explained in Chapter 5, an evaluation of the secondary chemistry has been done. The secondary system at PLGS typically operates on AVT chemistry treatment with morpholine and includes a full-flow condensate polishing plant, which was intended to be run continuously during system clean-up and plant restart. During the initial plant run-up, the morpholine chemistry was exchanged with ammonia to improve upon full-flow condensate polisher run times [Cook et al, 2014b]. This was even more considered to clean up the system after the discovery of organic contamination on the turbine rotors caused by an inadvertent incident that caused the low-pressure turbine rotors to be dropped into the Bay of Fundy.

Due to this permanent operation of the Condensate Polishing Plant (CPP), calculations have been made to compare:

- The cycle length of the CPP before breakthrough and rate of Flow Accelerated Corrosion (FAC) of carbon steel at various ammonia concentrations, Table 3-1;
- The cycle length of the CPP before breakthrough with ammonia or morpholine, Figure 3-3;
- The cycle length of the CPP before break of sodium related to the expected value at SG blow down, Figure 3-4, [Cook et al, 2014b].

It appears that if a high morpholine concentration is selected; the CPP cycle length is too short, explaining that the Utility selected ammonia treatment.

Table 3-1: Evaluation of CPP breakthrough and increased FAC risk on ammonia concentration in the condensate, after [Cook et al, 2014b].

NH ₃ (mg/kg)	Condensate pH _{at} 25°C	Amine break (hrs)	Boiler pH _{at} 25°C	Relative FAC risk (% vs. normal morpholine AVT) (model/chechworks)					
				BD	HTR 6 drains	RHD & SD	HTR3 drains	Extr. to HTR2	Extr. to HTR1
0.5	9.19	169.2	8.75	552	259	179	127	109	92
1.0	9.38	84.6	8.97	296/283.5	158/108.8	95/197.1	90/93.5	80/117.4	69/139.2
1.5	9.49	56.4	9.09	206.0	119	64	73	66	58
2.0	9.57	42.3	9.18	159/217.8	97/47.5	46/89.8	63/38.6	58/105.2	51/127.5
2.5	9.63	33.8	9.24	129.0	83	35	56	52	47
3.0	9.67	28.2	9.29	109/179.1	72/21.9	27/46.1	51/14.5	47/93.6	43/115.6
3.5	9.71	24.2	9.33	94.0	64	21	46	43	40
4.0	9.74	21.2	9.37	82.0	58	16	43	40	37
4.5	9.77	18.8	9.40	73.0	53	12	40	38	35
5.0	9.8	16.9	9.43	65/126	48/-3.9	9/10.9	37/9.7	36/80.5	33/99.2

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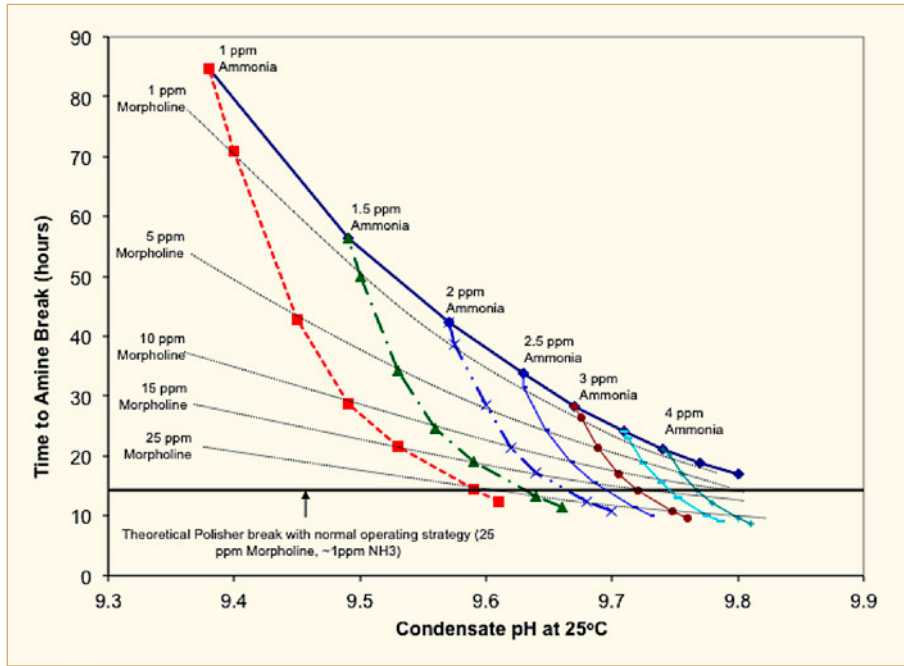


Figure 3-3: CPP breakthrough curves under various AVT chemistries [Cook et al, 2014b].

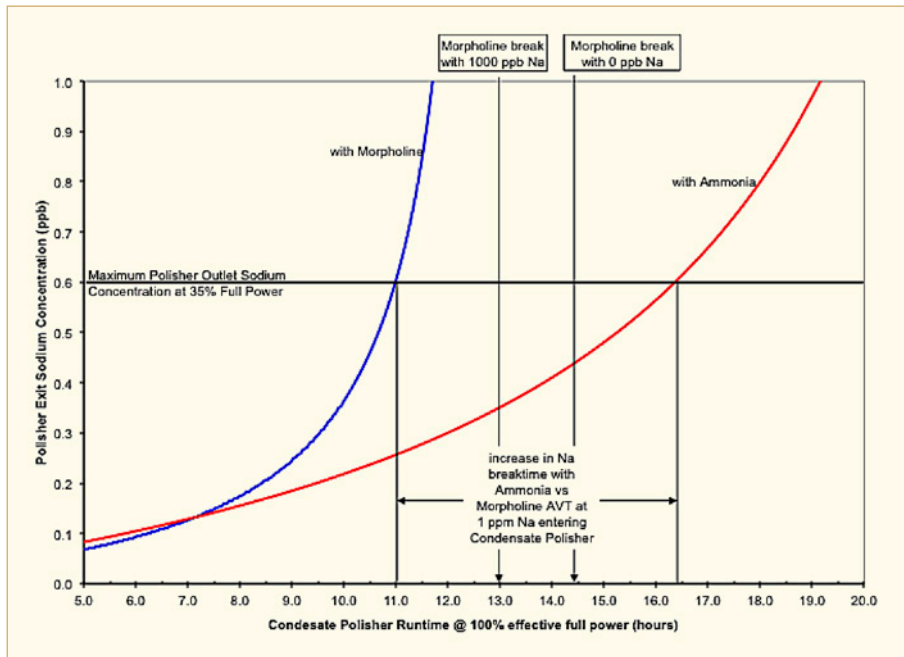


Figure 3-4: Comparison of polisher sodium break on ammonia versus morpholine [Cook et al, 2014b].

A team of American organizations compared experimental data to calculated value of activity coefficients that are used in MULTEQ™ code for the ionic strength dependence of equilibrium quotients for several ionisation reactions, including water itself.

4 Auxiliary systems, water purification, and waste

4.1 Chemistry of auxiliary systems

Ringhals unit 1(BWR) has taken measures to reduce or even stop the use of hydrazine in closed cooling water systems, hydrazine classification being harmful to the environment and hazardous for personal of the plant. Ringhals has to apply for a license every three years to continue the use of hydrazine. Consequently, a system to replace the use of hydrazine in closed cooling systems was installed in 2007 and includes three major steps of treatment. The first is to ensure low oxygen levels in the water using gas transfer membrane technology. The second is to control pH in the water by injecting potassium hydroxide (KOH) and finally, a mixed resin bed is used to clean up the anion and cation impurities in the water. The system has been connected to one of the turbine closed cooling system, earlier treated only with hydrazine. Possible material impacts from the new water environment have been evaluated using an autoclave system with coupons of relevant material. To this date, all three steps have worked out successfully and the autoclave material coupons show preliminary no or low general corrosion as expected due to the alternative water environment. The tests have continued with the objective of supporting a decision to fully remove hydrazine from the closed cooling water systems in Ringhals-1 [Hägg et al, 2014].

The main problems that can affect the operation of a cooling water system are linked to fouling or scaling of surfaces which, in turn, may cause a reduction in heat transfer or increase the risk of corrosion. In cooling water systems operating with natural waters, some fouling and scaling occur naturally. VTT Research centre in Finland studied the effect of ethanol, sodium hydroxide, phosphoric acid and phenol as well as rapeseed oil on fouling, heat transfer and corrosion of heat exchanger material (titanium) in synthetic brackish water simulating Baltic Sea water. The chosen chemicals are among the most frequently transported chemicals (rapeseed oil as a representative of oily products) on Baltic Sea, which may thus be present in the cooling system. The study has been done with a specialized flow-through cell, with on-line monitoring of a heated sample by methods such as electrochemical impedance spectroscopy and linear polarisation resistance. The corrosion rates of titanium plates exposed to ethanol, sodium hydroxide, phosphoric acid, phenol or rapeseed oil solutions with concentrations of 0.1, 1.0 or 10.0ppm are not greatly different from corrosion rates of titanium plates exposed to artificial brackish sea water, and therefore a chemical spill near a nuclear power plant is likely to cause no increase in the corrosion of titanium in the cooling water system, as can be seen on Figure 4-1. It seems also that the chemicals have not a marked effect on the heat transfer rate or that they actually increase the rate of heat transfer slightly [Velin et al, 2014b].

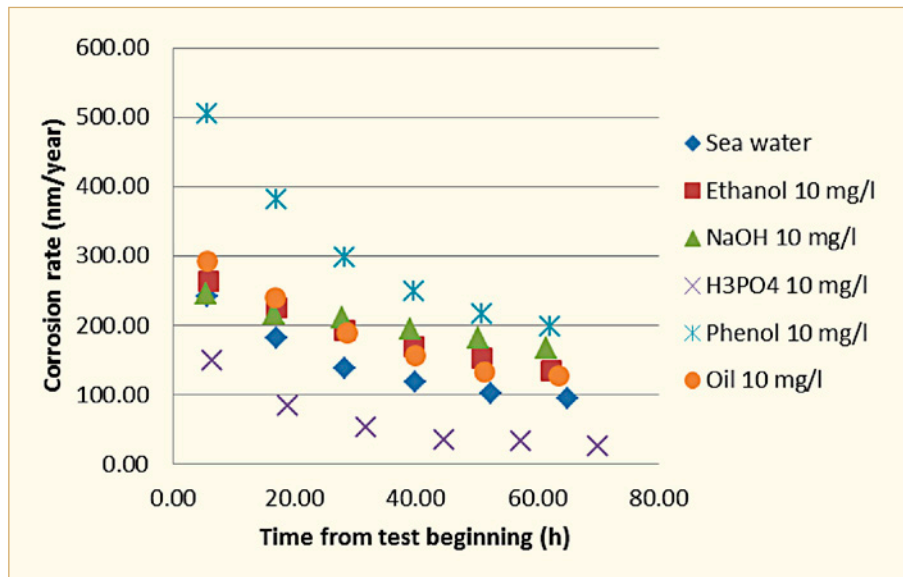


Figure 4-1: Corrosion rate of titanium when exposed to chemicals with concentration of 10 mg/l added in sea water. T = 80°C [Velin et al, 2014b].

EPRI developed a second revision of the Closed Cooling Water Chemistry Guideline that addresses the use of chemicals and monitoring methods to mitigate corrosion, fouling, and microbiological growth in the closed cooling-water (CCW). The paper does not include the recommended values of this guideline. One of the objectives was to get consistent limits, which may differ from vendor to vendor. For 300 series stainless steels, impurity limits for chloride and fluoride are established at 10ppm each. No other value has been published at the conference [McElrath & Breckenridge, 2014]. Another EPRI guideline is related to Open Cooling Water Chemistry.

4.2 Primary coolant purification and decontamination

An EPRI benchmarking compared various PWR data of Ion Exchange Resins (IER) for the RCS purification with the following observations [McElrath et al, 2014].

- For a PWR, the highest DFs and removal efficiencies are obtained during clean up from the forced oxidation step during shutdown to a refuelling outage. The forced peroxide oxidation step can generate ^{58}Co concentrations between 1 and 10 Ci/g (3.7 E10 to 3.7 E11 Bq/g) in the reactor coolant. As a result, it is not uncommon for a PWR to report ^{58}Co removal efficiencies during this time period >99.9%.
- PWRs generally use layered beds during shutdown operation and mixed beds during power operation.
- PWRs have focused on the use of macro porous anion resin as an overlay in the CVCS deep beds in order to improve the removal efficiency of colloidal activated corrosion products during shutdown. Where initially some plants focused only on using the macro porous anion resin overlay for shutdown operation, more plants now are using a macro porous anion resin overlay during power operation.
- A number of PWRs (particularly those using a macro porous anion resin overlay in the CVCS demineralizers) intentionally bypass filters upstream of the CVCS deep beds in order to eliminate the dose associated with changing out filter cartridges. The thought process is to use the macro porous anion resin overlay and cation resin middle layer for particulate (colloidal) filtration. This should result in less fouling of the bottom mixed bed resin layer.

- Braidwood Unit 1 and Unit 2 data showed no significant performance differences between shutdown beds that used the NRW5070 overlay and shutdown beds that used the modified cation resin. Both resins produced ^{58}Co removal efficiencies $>99.9\%$ during shutdown. Braidwood has used sub-micron (0.05 micron) filter cartridges in their post demineralizer filter vessels during power operation. The station had not experienced any significant pressure drop issues with the smaller size filters and has reported some improvement in some radiation protection metrics.

The main Indian Laboratory for NPP developed a material to adsorb antimony. The purpose is to avoid redeposition of radioactive antimony after decontamination for eliminating ^{60}Co . Six commercially available sorbents and one synthesised high performing sorbent (TA-Chitosan beads) in the form of stable beads, using nano- TiO_2 and chitosan, have been compared for their antimony sorption properties. Tulsion A33 is good for removing Sb(V) while titania based materials are superior if both Sb(V) and Sb(III) are present. Cross linked Chitosan TiO_2 composite beads prepared were shown to be efficient antimony sorbent for removing both Sb(V) and Sb(III) forms, in the absence of ferrous ions. Radiation stability of the cross linked Chitosan- TiO_2 beads for its use in nuclear industry was established [Nishad et al, 2014].

EDF investigated the frequency of replacement of the Filters used for solid particles removal from the primary system in the fleet of 58 reactors. The primary system with its connected circuits is shown on Figure 4-2.

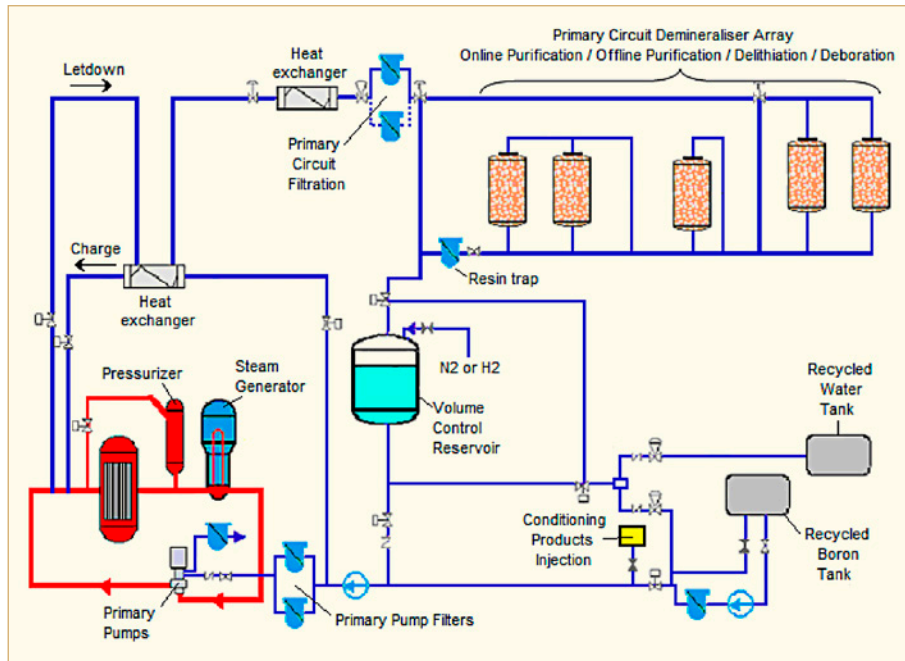


Figure 4-2: Simplified diagram of the RCV/CVCS system for 1300 MWe/N4 reactors [Mascarenhas et al, 2014].

Indeed, since 2008, a steady increase in the consumption of filters has been noticed, and therefore an increase in the amount of solid waste to treat, as highlighted on Figure 4-3.

5 Lifetime management, plant ageing and modernisation

Following a refurbishment outage lasting 4.5 years, the CANDU 6 reactor at Point Lepreau in Canada, was restarted. As much of the Primary Heat Transport system was replaced during the refurbishment, particular care to chemistry control in this system was required during its return to service, in particular the completion of a hot conditioning process to form an adherent magnetite layer on the carbon steel components was carried out, for ensuring reducing conditions. This corrosion of the carbon steel during the hot conditioning process has been monitored with an on-line deuterium effusion probe, HEPro™, for the first time in a CANDU plant. This additional information provided significant confidence of successful implementation of the restart strategy to the chemistry team and allowed the success of the conditioning process to be verified before proceeding to increased power operation [Stuart et al, 2014].

In addition to the replacement of the zirconium alloy pressure tubes, 380 fuel channels, calandria tubes and end fittings, and the 760 associated feeder pipes that constitute the reactor core, the secondary system of CANDU 6 reactor at Point Lepreau, 680 MWe, first commercial operation in February 1983, was also refurbished after 25-30 years of operation. Considerable work was undertaken, including a turbine upgrade to increase the power output by 3-5%, a complete re-tubing of the titanium condensers, disassembly and cleaning of the steam reheaters and many other minor upgrades and repairs. The plant was returned to service in November 2012, more than three years behind the original schedule [Cook et al, 2014b].

In the perspective of prolongation of Borssele (PWR) operation until 2034 (20 additional years) in Netherlands, it has been decided to modernize the sampling system for the steam-water cycle. Figure 5-1 gives an overview of the treatment and surveillance study [Drexler et al, 2014b].

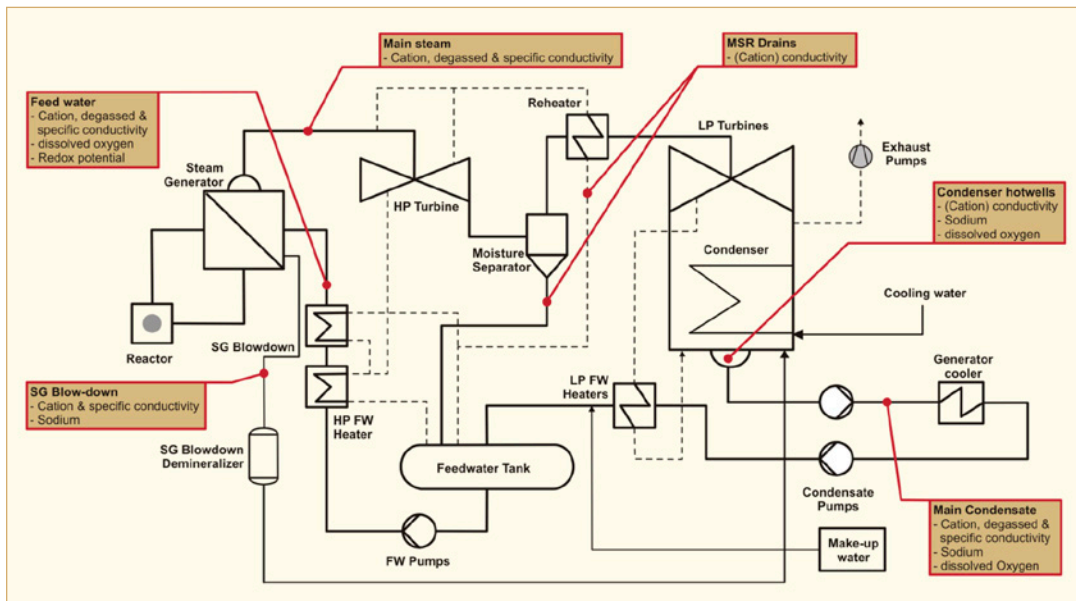


Figure 5-1: Steam-water cycle of NPP Borssele with main on-line instrumentation according to the concept study [Drexler et al, 2014b].

The French Utility (EDF) has its own system of performances evaluation, the Nuclear Inspectorate, which is independent and makes its own audit in addition to what is done under WANO or IAEA. These inspections include the field of chemistry and showed that the specifications are correctly followed and the various areas of improvements for safety and plant life duration have been examined [Jacquier, 2014].

EDF Energy intends to build two EPR units at the Hinkley Point C site and has initiated the studies to build two more EPR units at Sizewell C in UK. Best Available Techniques are studied with some international feedback in addition to EDF experience and support.

6 Conclusions

The NPC Conference on chemistry is the major event with real international information covering most countries, all types of reactors, with the point of view from Manufacturers, Utilities, Operators, Research Laboratories, Regulators, various vendors. This conference, held every other year, took place previously in Paris in September 2012, the last one in Japan, Sapporo, last week of October 2014, and the next one will be in UK, Brighton, on first week of October 2016.

This conference deserves a large coverage and two separate reports are giving the most relevant information of NPC 2014: this report for PWR, VVER, CANDU, FBR and some common systems or topics, while another report is covering BWR, Fukushima accident and also some common systems.

This report also briefly covers a one-day French workshop on the relation between chemistry and the environment.

6.1 Primary coolant

The trend is always to try operating with the optimum pH_T in the primary coolant of PWR for mitigating Operational Radiation Exposure (ORE). However, with the fuel of high performances, it is not always feasible to reach this optimum pH at the beginning of fuel cycle without the use of Enriched Boric Acid (EBA) that has been selected for the new EPR. Otherwise, a too high lithium concentration would not be acceptable for the fuel and other materials, while anyhow, some studies also showed the detrimental impact of high boron concentration on fuel crud. EBA is also considered in Eastern Europe for VVER.

Many laboratories are still studying the mechanisms of corrosion products transport in the primary coolant for dose rates mitigation and for understanding the crud formation on the fuel cladding. There are also many research studies on crud deposition on the fuel and the related resistance to corrosion of fuel cladding.

With the progressive replacement of some components (mainly SG) with the sensitive Alloy 600, most of the organisations are thus focusing on other issues. However, the USA, with still many plants in operation with Alloy 600, is continuing to look at remedies for PWSCC, with higher Dissolved Hydrogen content and zinc addition.

Zinc injection in the primary coolant should be beneficial at least for thin materials such as SG tubes with Alloy 600, and this is the reason for its selection in some units like the French ones with Alloy 600 TT SG tubes. The number of PWR units adding it for dose rate mitigation is increasing despite the results that are sometimes clearly highly beneficial (Japan) and some other times not so clearly (France), but at least never detrimental. Zinc option is also included in the new design of VVER units. Its early application at Tomari, from the Hot Functional Testing (HFT), gave good results, together with other improvements. Most of the studies are claiming that zinc addition decreases the uptake of radioactive corrosion products on components surfaces.

There are continuous investigation on manufacturing process and HFT to optimize the metal surfaces and thus decreasing further dose rates, since this appears to be a very efficient way of reaching very low values during future operation, while other approaches already almost reached their optimum. However, HFT process is obviously only a solution for new plants in a limited number of countries.

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Nomenclature

AOA	Axial Offset Anomaly
ASCA	Advanced Scale Conditioning Agent
BARC	Bhabha Atomic Research Centre (India)
CANDU	CANada Deuterium Uranium type of Reactor
cc/kg	cubic centimetre (per kg): used for H ₂ concentration in RCS = ml/kg (under normal pressure and temperature)
CCW	closed cooling-water
CEA	Commissariat à l'Energie Atomique / French Atomic Energy Commission
CGR	Crack Growth Rate
CHC	Critical Hydrogen Concentration
CILC	Crud Induced Localized Corrosion
CIPS	Crud Induced Power Shift
CPP	Condensate Polishing Plant
CRUD	Chalk River Unidentified Deposits
CVCS	Chemical and Volume Control System
DMA	Dimethylamine
DH	Dissolved Hydrogen
DMT	Deposit Minimization Treatment
EBA	Enriched Boric Acid
ECP	Electro Chemical Potential
ECT	Eddy Current Test
EdF	Electricité de France
EPR	European Pressurized Water Reactor
EPRI	Electric Power Research Institute
ETA	Ethanolamine
FAC	Flow Accelerated Corrosion
FBR	Fast Breeder Reactor
FFA	Film-Forming Amine
GWTS	Gaseous Waste Treatment System
HFT	Hot Functional Test
HPWR	Heavy-water Pressurized Water Reactor
IAEA	International Atomic Energy Agency
ICP	Inductively Coupled Plasma - Optical Emission Spectrometry (ICP-OES or ICP)
ICP-AES	Induced Coupled Plasma - Atomic Emission Spectrometry
IER	Ion Exchange Resins
IGSCC	Intergranular Stress Corrosion Cracking
KWU	Kraftwerk Union (Nuclear Power Plant Section of Siemens)
LPR	Long-Path Recirculation (of dispersant)
LTU	Online Long-Term Use (of dispersant)
MA	Mill Annealed
MHI	Mitsubishi Heavy Industry
MOX	Mixed Oxide (Fuel elements)
MVP	Mechanical Vacuum Pump
NDE	None Destructive Examination
NPP	Nuclear Power Plant
ORE	Occupational Radiation Exposure
PAA	Poly Acrylic Acid (dispersant)
PVHWR	Pressurized Vessel Heavy Water Reactor
PWR	Pressurized Water Reactor
PWSCC	Primary Water Stress Corrosion Cracking
RCP	Reactor Coolant Pump
RCS	Reactor Coolant system
RFO	Refuelling Fuel Outage
RPV	Reactor Pressure Vessel
SCC	Stress Corrosion Cracking