

# LCC-1 Annual Report

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## 1 INTRODUCTION (STIG SANDKLEF)

Nuclear energy is at a turning point. Again!

But this time it may be for real. The US National Energy Policy of 2001 has a vision that by 2020 20 000 MWe shall be added to the grid. To support this vision legislation to help finance the first three reactors of next generation has been passed, and a simplified licensing structure is in place. A couple of utilities are in the process of applying for a COL.

A similar picture is developing in several other countries such as China with perhaps 40 000 MWe of nuclear capacity added until 2020, France has decided that the capacity of her 55 nuclear units will be replaced by new nuclear units, starting in 2015. Finland is constructing the first 1500 MWe EPR, Japan has confirmed its long term strategy to maintain a certain share of nuclear energy in the energy mix and the list can be made longer.

There are several reasons for this development where the safe and reliable operation worldwide of nuclear plants in the last 19 years perhaps is the most important. Strategically; climate change, energy security and environmental benefits of nuclear energy play a large role.

For a utility about to make an investment decision the most important parameters are safety, reliability and production cost. Also here we have a positive trend as illustrated in Figure 1-1 where the competitiveness of nuclear energy is clear. With todays prices of fossil fuels the differences should even be much larger.

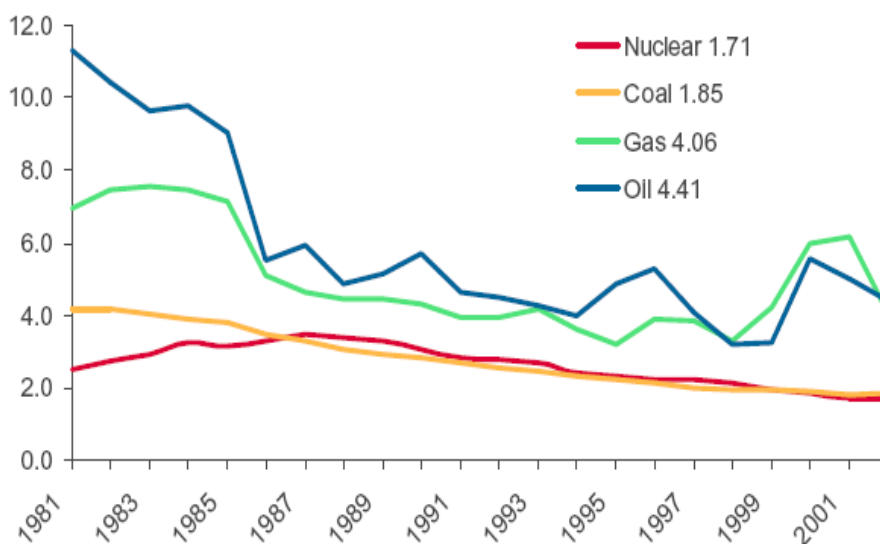


Figure 1-1: US Electricity Production Cost (1981 – 2002). In 2002 cents per kilowatt-hour, Jones, 2004(b).

The largest threats to safe, reliable and cost effective operation of nuclear plants have historically been related to various types of corrosion and materials problems. Even today these problems are the most costly issues experienced by operators of nuclear plants. Well known examples are the stress corrosion cracking of stainless steel piping in BWRs and corrosion damage to steam generators in PWRs. Especially, when corrosion cracking has affected pressure retaining components or core support structures safety concerns are important.

It is vital that these threats to the successful operation of nuclear plants are minimized as much as possible and major improvements have also been made over the last 25 years.

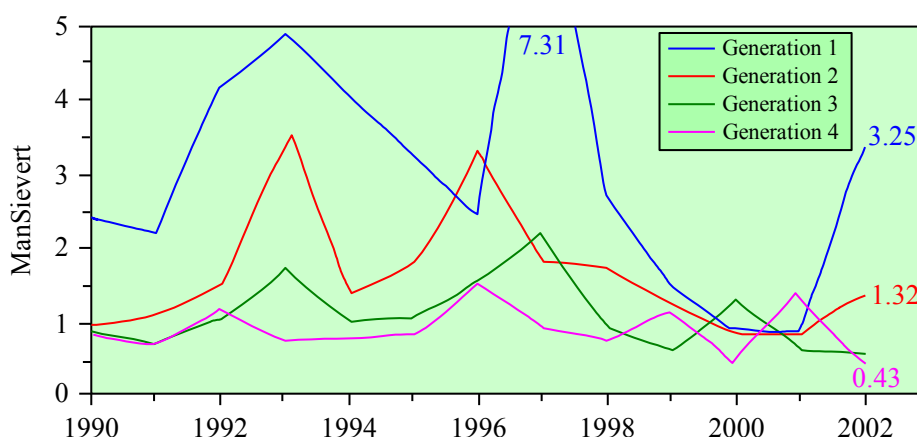


Figure 1-2: Swedish BWRs Collective Radiation Exposure, modified figure according to KSU 2003.

An illustration of the development of water chemistry control and materials selection is given in Figure 1-2 where collective radiation exposure is given for the Swedish BWRs. Generation 1 was designed in the 1960ies and is still affected by non optimal materials in the primary circuit even if increased knowledge and skills in water chemistry management gives a trend of gradual improvement. Generation 4 (designed in the early 1980ies) starts with much better materials selected and coupled with more knowhow of water chemistry shows a much better performance.

The skill and competence of the people responsible for water chemistry and materials in the plants is thus of great importance for successful operation. As with practically all other categories of skilled people in the nuclear industry there is now an ongoing shift of generation where new specialists are recruited with good education but not always with access to history and past experience in their fields of responsibility.

The analysis of this situation has led ANT International to launch the LCC program as one way of assisting nuclear utilities in meeting these challenges in a most effective way.

The overall objective of the LCC program is to enable nuclear utilities to:

- Gain increased understanding of reactor water chemistry related to successful plant operation and a continued integrity of RCS materials while keeping radiation exposures low.
- Assist in the training and education of a new generation of chemistry and materials experts at nuclear utilities.
- Establish a new independent (free from big organisations and vendors) meeting point for utility experts to enable free and critical discussions and experience exchange.

The objective is met through critical review and evaluation of recent data, identification of important new information and discussion of its significance in relation to water chemistry and put in perspective to how today's situation has evolved.

The evaluations are based on large amounts of non-proprietary data presented at technical meetings and published in the literature.

This report covers the following:

- International Conference on “Water Chemistry of Nuclear Reactor Systems”, Oct. 11-14, 2004 San Francisco.
- 6<sup>th</sup> International Seminar on Primary and Secondary Side Water Chemistry of Nuclear Power Plants, Budapest, 2005.
- Symposium of Environmental Degradation of Materials in Nuclear Power Systems-Water Reactors, 2005, Salt Lake City.
- Symposium on Water Chemistry and Corrosion of Nuclear Power Plants in Asia, Kyungju, Korea, oct. 2005.
- VGB Conference – Chemie im Kraftwerk- oct 2005, Mainz.

Journal publications are being monitored through several literature searches of worldwide publications and the important papers are summarised and critically evaluated. This includes the following journals:

- Nuclear Engineering and Design
- Kerntechnik
- Power Plant Chemistry
- Water Chemistry Guidelines (if and when published)
- IAEA documents



The large collective experience gained by the reviewers and authors of this report in past and current projects is an important factor in making the evaluation, ensuring that the compiled information is put in perspective and that the most important information is emphasized.

The report is structured in the following sections:

- Introduction
- Coolant Quality and Control Issues
- Materials Selection for the Primary Circuit
- Primary Circuit Corrosion
- Dose Rate Build-up and Control
- Fuel/Water Chemistry Interaction
- Current Issues and Material Developments

In particular the third section is a very comprehensive review of materials selected for the primary circuit, going back to early designs and successive developments to facilitate the understanding of today's situation.

Each section will be presented by the respective author in a one day seminar.

## **2 COOLANT QUALITY AND CONTROL ISSUES**

Corrosion control is imperative in reactor coolants for optimum performance of the plant. Without it, system integrity may be jeopardized, activity transport may give rise to unacceptably high doses of radiation to plant staff and corrosion product build-up may create barriers to heat transfer and interfere with the smooth operation of mechanical components.

Corrosion control is dependent upon the chemical parameters of the coolant. As nuclear industry has developed, the knowledge of controlling corrosion using water chemistry has also progressed. As examples, activity transport is controlled in PWRs by operating the coolant under higher pH-values and by injecting Zinc. Oxidizing conditions, that promote stress corrosion cracking in BWRs are being counteracted by hydrogen dosing and noble metal injection.

Nevertheless, corrosion problems continue to occur. In addition economic pressures are leading to more rigorous operating conditions in power reactors. Fuel burn-ups are to be increased, higher efficiencies are to be achieved by running at higher temperatures, plant lifetimes are to be extended, and an increasing number of reactor systems are load following. Constraints such as these impose greater demands on the system and compel the reactor chemists to acquire more information to keep corrosion under control.

## 2.1 PWR WATER CHEMISTRY (ROLF RIESS)

The reactor coolant system, RCS, in PWRs consists of 2 to 4 heat transfer loops connected to the reactor vessel. Figure 2-1 is a schematic overview of a 4-loop system. Each loop in the arrangement shown contains one steam generator and one reactor coolant pump. The RCS also includes the pressurizer, connected to one of the reactor vessel outlet pipes, with attached safety valves and power operated relief valves to provide protection against over-pressurization. These valves discharge to a quench tank which cools and condenses the steam.

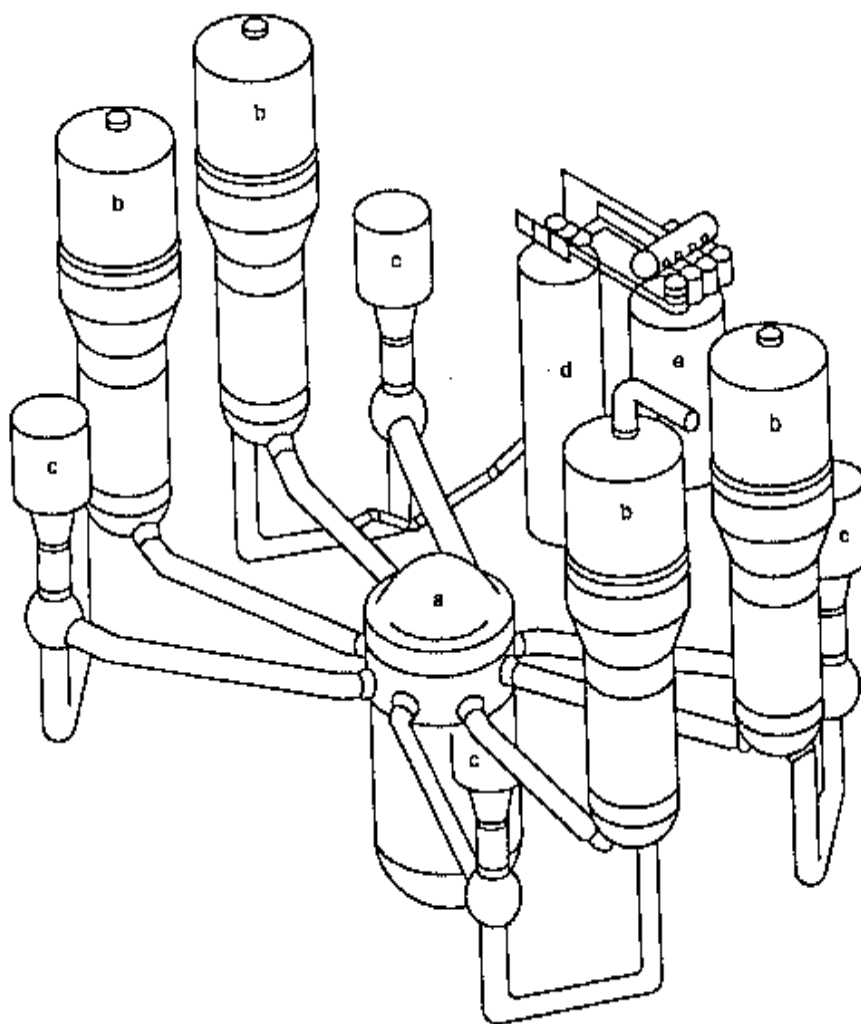


Figure 2-1: Schematic overview of a 4-loop reactor coolant system: a, reactor vessel; b, steam generators; c, reactor coolant pumps; d, pressurizer; e, pressurizer relief tank, Courtesy of FANP.

The primary coolant in the RCS serves as a moderator and is a medium for transporting heat from the core to the steam generators. Hence, it must not endanger plant operation by the corrosion of materials and consequences thereof. The task of water chemistry can be divided into the following main points:

- Metal release rates of the structural materials should be minimal.
- The occurrence of localized forms of corrosion should be counteracted.
- The transport and deposition of corrosion products must be influenced in such a manner, that contamination of the primary coolant system is kept low.
- The deposition of corrosion products on heat transfer surfaces, particularly on fuel assemblies, should be prevented as far as possible.
- Radiolytic formation of oxygen should be suppressed.

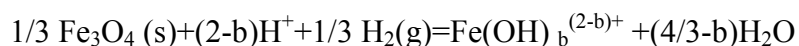
In certain instances, situations may be encountered where chemistry conditions that are optimum for achieving one goal can lead to a decreased level of achievement relative to other goals. As a result of such considerations the water chemistry specifications must define parameters to achieve a balance among the five goals, recognizing that highest priority is designed to materials and fuel integrity goals. Although the other goals are in the second line of priority, like radiation build-up, can not be ignored.

The water chemistry conditions are also requiring the proper selection of the materials which are in contact with the primary coolant, see section 3 for more details. They are:

- Austenitic stainless steels of components and piping of the primary system
- Zirconium alloys for the cladding of fuel assemblies
- Incoloy 800, Inconel 600 MA or TT; Inconel 690 TT for steam generator tubes. Stainless steel tubing is used in VVER SGs.
- High alloy materials (ferritic stainless steels) of low surface area for internals of the primary system.

The water chemistry conditions applied to these materials must fulfil the above mentioned chemistry requirements. Thus the primary coolant of PWRs, which contains boric acid (900 – 1800 ppm B at Beginning Of Cycle, BOC) as a neutron absorber is chemically conditioned by the addition of isotopically pure Lithium (Lithium-7) hydroxide (2 – 5 ppm Li at BOC) as a non-volatile alcalizing agent and by the addition of hydrogen.

Historically, the starting point for all discussions about the correct pH in PWR Primary Coolant can be found in P. Cohen's book "Water Coolant Technology of Power Reactors", 1969, and especially in the Chapter "The Physical Chemistry of Water and Aqueous Solutions". The central point in this document is the work of Sweeton et al., 1968, who have reported measurements of the solubility of Fe from  $\text{Fe}_3\text{O}_4$  and they wrote the general dissolution reaction as



This equation is valid for the temperature range of interest and applicable to dilute acidic and basic solutions. It also became clear that isotopically pure Lithium-7-hydroxide is the most suitable pH control agent to be used in the PWR Primary Coolant.

The result of Sweeton's work can be seen in Figure 2-2.

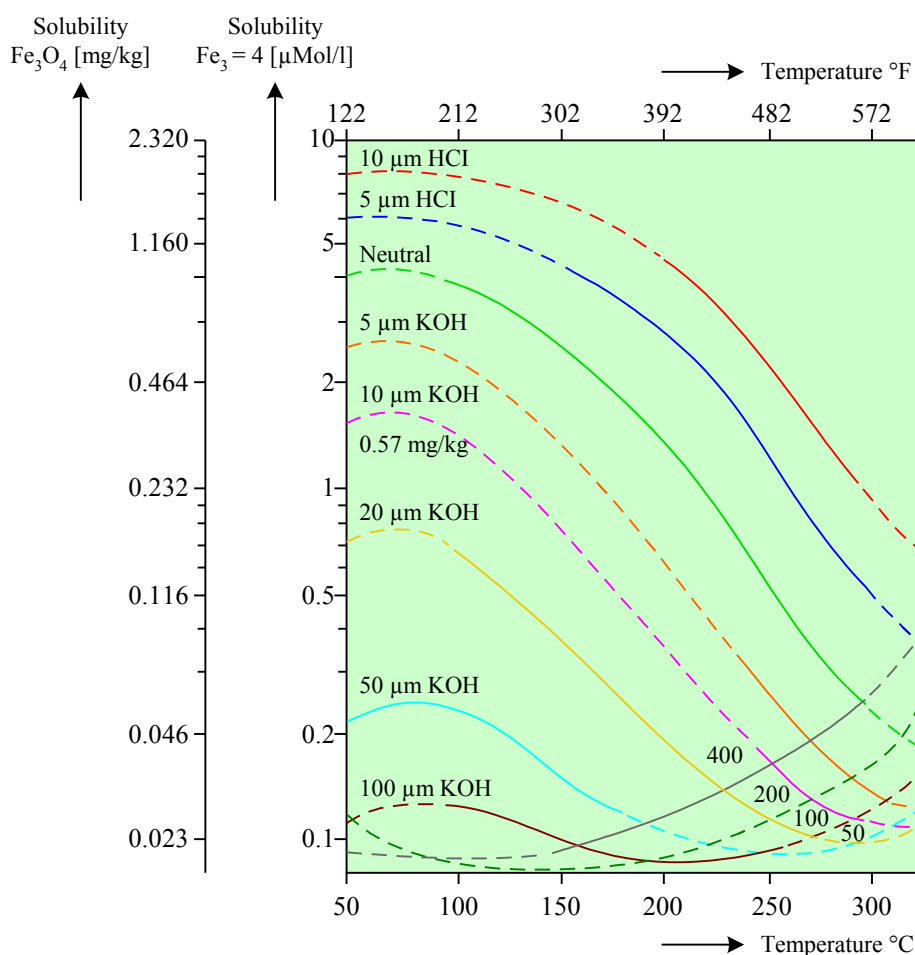


Figure 2-2: Magnetite Solubility by Sweeton et al., 1968.

These data suggested, that under the conditions of a PWR Primary System the optimum pH should be pH 6.9 at 300°C. At these conditions iron solubility is at a minimum and thus the transport of iron based crud should also be a minimum. Furthermore, 2 ppm of Lithium were sufficient to achieve a pH of 6.9 at BOC ( $\approx 900$  ppm B for an annual cycle). These 2 ppm Lithium were also considered to be low enough to avoid any corrosion attack on the fuel elements.

However, later on it was recognized, that the contribution of Nickel is much more important to the primary side corrosion product inventory than the iron. Further on it was found, that Nickel ferrite is a major constituent. Consequently the solubility behaviour of Nickel ferrite was investigated and it was found, that a pH of 7.4 should be the solubility minimum. However, a pH of 7.4 could not be adjusted at BOC since 2 or 2.2 ppm Lithium was at the upper specified limit in order to prevent Lithium induced corrosion of the fuel element cladding. As a consequence, the first two thirds of a cycle the 2 ppm were kept constant till reaching the pH of 7.4 and then Lithium/Boron ratio was adjusted to stay at 7.4 till end of the cycle.

In order to discuss this transition of the last 35 years it may also be useful to look in parallel to the current Lithium specifications as they are applied in various countries, see Table 2-1. A second question is, whether these data are describing the development in the relevant time period.

Table 2-1: Lithium Specifications as they are valid in various countries.

Lithium Specification in ppm	
Germany	0.1 - 2.1
Japan	0.2 - 2.2
France	0.4 - 2.2
USA	-- (plant specific programs)
Czech Republic	<0.35 mmol/l $\Sigma$ K, Li, Na

At a first glance there seem to be little or no changes in the Lithium specification compared to the early days of commercial PWR operation. Also between the different countries negligible differences do exist, except USA, where plant specific specifications have to be applied and where new B/Li-strategies were developed.

To say it in other words: The specified Lithium values are not very helpful to understand the worldwide Lithium strategies. It is much more important to look after the Lithium/Boron ratio because changes in the Li/B control were experienced by the industry, especially in the US, establishing specific modes of operation.

To explain these changes, it is convenient to use the information shown in Figure 2-3. This figure shows, as a first more precise interpretation, the relation between Lithium and Boron in the coolant. This relation is handled in such a way, that the pH at temperature is 6.9 which is called **coordinated Lithium/Boron chemistry**. It means, that the  $\text{pH}_T$  is constant over the entire cycle.

A second mode is the **modified Lithium/Boron chemistry**, where at the BOC in annual cycles a concentration of 2 – 2.2 ppm Li is used and kept constant till reaching a desired pH of e.g. 7.4 and then the pH 7.4 line is followed by the appropriate Li/B-coordination (see Figure 2-3, lower part and Boron values of <1000 ppm).

In case of extended fuel cycles, the Lithium concentration at BOC can be higher, e.g. in order not to fall below a  $\text{pH}_T$  of 6.9. The chemistry will be operated in a coordinated mode of 6.9 till reaching a Lithium level of 2.2 ppm and will then follow the above description.

The third alternative is to operate with an **elevated Lithium/Boron Chemistry** where a level of 3.5 ppm Li is used at BOC till reaching a pH of e.g. 7.1. At this pH the Li/B coordination is adjusted to stay at pH 7.1 till the end of the cycle.

The fourth alternative is the **constant elevated Lithium-Boron Chemistry** where at the BOC a high Li-level of e.g. 5 ppm is adjusted and during the cycle a Li/B-coordination keeps the pH constant throughout the cycle. This case is identical with the first one except that the  $\text{pH}_T$  is adjusted at a higher level.

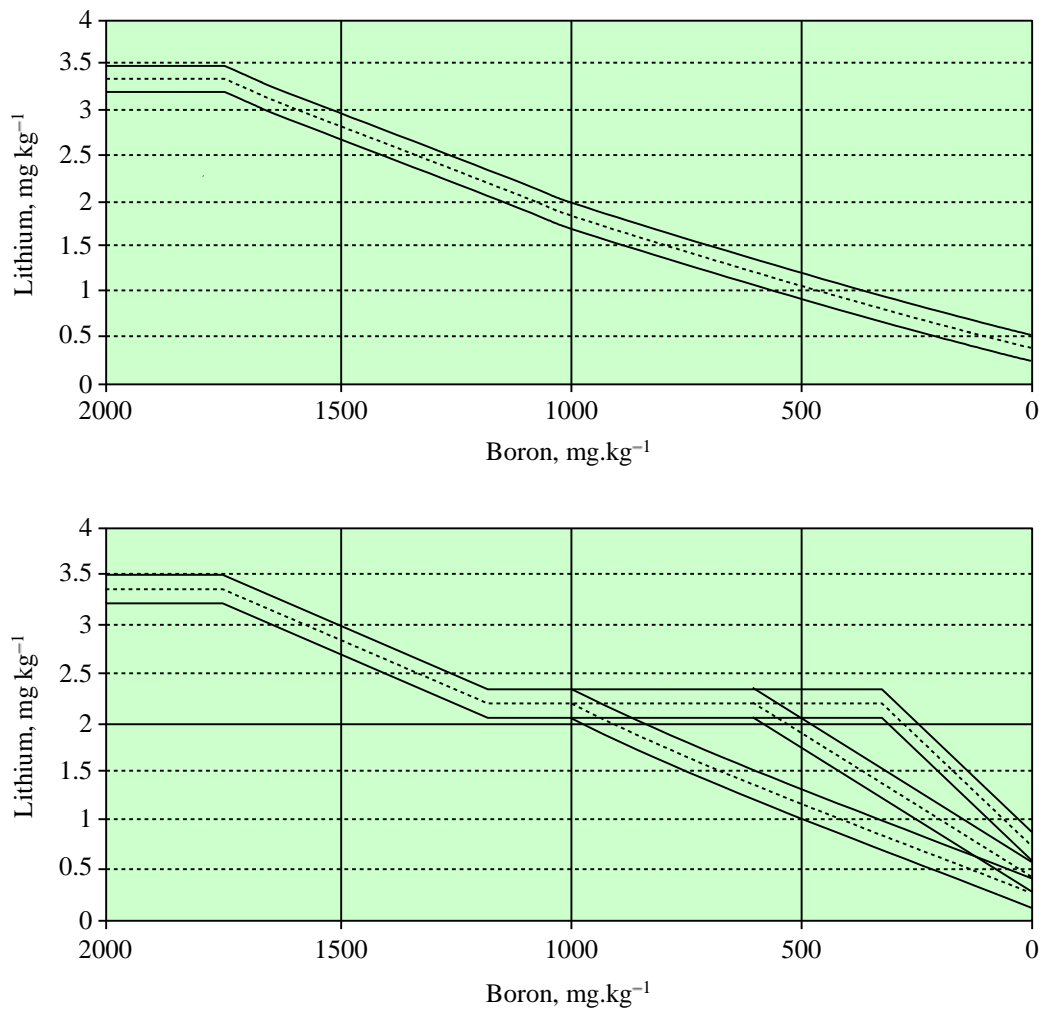


Figure 2-3: Top figure: Co-ordinated (6.9) and bottom figure: Modified (7.0, 7.2, 7.4) Chemistries, modified figure according to Garbett, 2003.

During all optimisations (new mode) of the PWR primary coolant specifications a permanent conflict existed between increasing the pH e.g. due to radiation field consideration or prevention of crud accumulation and the fear of Zircaloy corrosion caused by the Lithium concentration mechanism, see Figure 2-4.



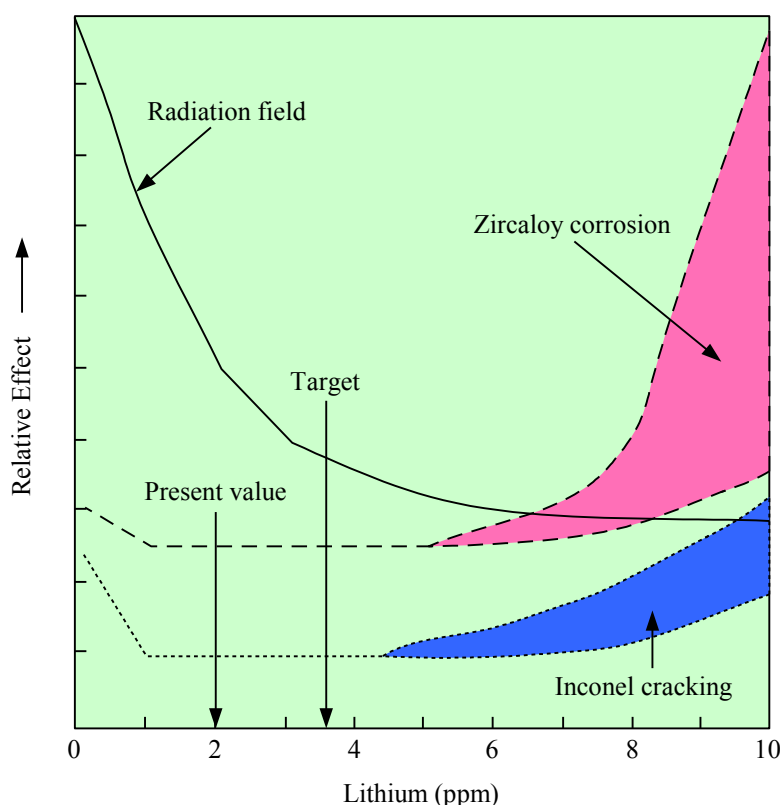


Figure 2-4: PWR Primary Coolant Chemistry Effect of Lithium Concentration (Schematic Diagram), modified figure according to Riess & Millet, 1994.

The desire to increase the Lithium-concentration to be applied to the primary coolant requires higher target concentrations as already reported by Riess & Millet, 1994. Such modifications however, implied the risk of Zircaloy corrosion and Inconel cracking. Despite these risks, the **modified and elevated Lithium/Boron chemistry** were recommended specifically in many US PWRs. But still the radiation field problems and Axial Offset Anomaly, AOA, in plants with high duty cores stimulated to go to even higher Li-concentrations.

The Lithium specification as shown in Table 2-1 on one side and the Lithium/Boron concentration modes on the other hand need some more country specific discussion. For example, the time frame for chemistry changes in the US is shown in Figure 2-5, Turnage, 2004.

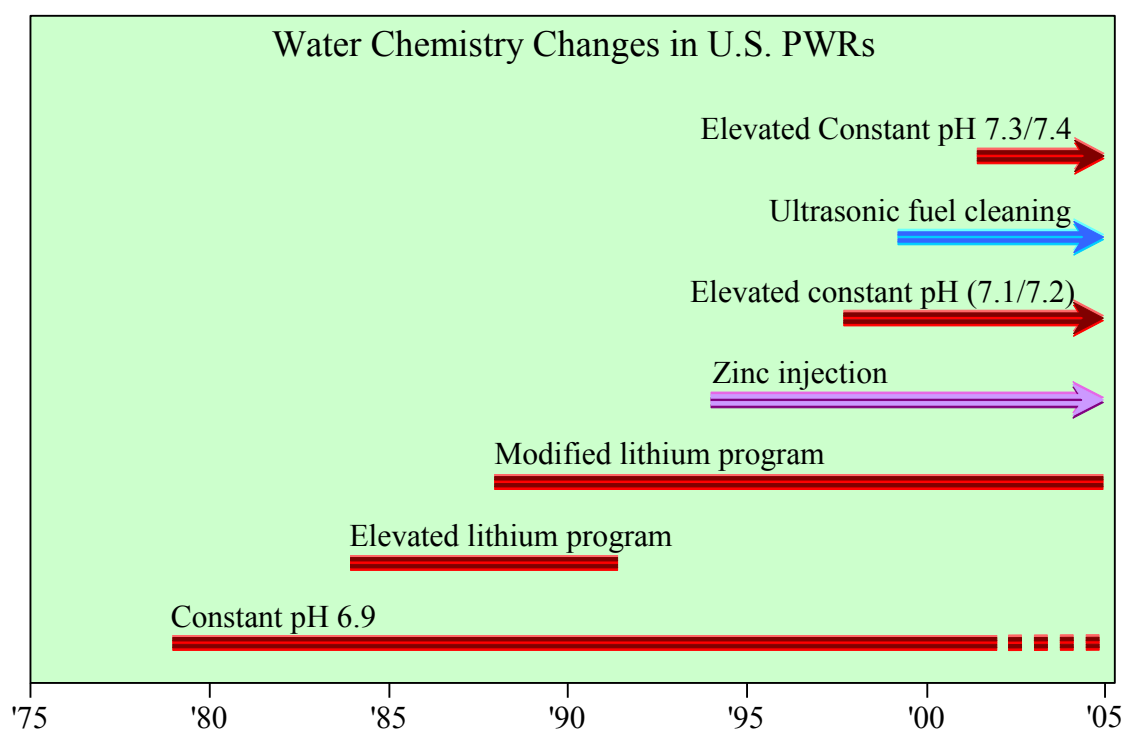


Figure 2-5: Water Chemistry Changes in U.S. PWRs, modified figure according to Turnage, 2004.

Regarding the application of all the LI/B-chemistries in non-US operating PWRs there are some differences applied in the various regions and countries. Such variations depend also on the dates of the application. In the early 1970s worldwide the Lithium concentration was operated between upper and lower specified values. The first step to the coordinated chemistry was made in the late 70s/early 80s in order to reduce crud deposition on fuel rods and to minimize radiation fields. The selected  $\text{pH}_T$  was 6.9, which was also consistent with the  $\text{pH}_T$  for iron solubility minimum under PWR primary side conditions.

In the mid 1980s, many PWRs introduced the modified chemistry based on operational experience (see above), because at this time high and constant Lithium as a pH strategy produced better result than the coordinated chemistry, as far as crud deposition and radiation fields are concerned.

The German PWRs are still operating today with this type of chemistry and therefore the specification, given by FANP GmbH and VGB respectively, have an upper Lithium limit of 2.1 ppm. Only in very specific cases, utilities can apply Lithium values higher than 2.1 with the consent of the fuel manufacturer.

In the second half of the 1980s in Ringhals an elevated Lithium strategy with an initial concentration of 3.5 ppm was tested, however, this project was stopped due to concerns for corrosion of Inconel 600 steam generator tubing and Zircaloy-4 cladding corrosion. Despite this kind of “Warning”, several US PWRs increased the cycle length and thereby the Boron concentration at BOC to values of  $\approx 1.500$  ppm Boron. The motivation for this step is described above. According to the general understanding the  $\text{pH}_T$  of  $>6.9$  can only be maintained when higher Lithium concentrations are allowed. This became operational practice by starting the cycle operation with 3.5 ppm Lithium, which means an elevated chemistry as described above.

### 2.1.1 Definition of Terms

The above mentioned modes/regimes of operation are described in “Guidelines”, as for example:

- In the US the EPRI PWR Primary Water Chemistry Guidelines which are issued every 4 years and which have their fifth revision dated 2003. Proprietary document; not available to the author.
- In France the “Specifications Chimique des Centrales RET” which are updated on a non regular basis.
- In Germany the “Richtlinie „R 401”” which is the successor of an earlier guideline – VGB – R 401 J- released in 1988.
- In Russia: See publications as discussed later

The purpose of the guidelines is to describe the **Quality Requirements** of the reactor coolant. However, it has to be distinguished between various operational modes, see Table 2-2. As far as the various Guidelines provide information to this or other issues, they will be compared in this document.

Table 2-2: Operational Status Modes.

	EPRI	VGB	EdF
Cold shut-down	$<250\text{ }^{\circ}\text{F}$	$<120^{\circ}\text{C}$	$<120^{\circ}\text{C}$
Start-up	$>250\text{ }^{\circ}\text{F}$ But not critical	$>120^{\circ}\text{C}$ But not critical	$>120^{\circ}\text{C}$ But not critical
Power operation	Reactor critical	Reactor critical	Reactor critical

### Typical Values

Typical values are values, which can be achieved during undisturbed steady-state plant operation. They are plant specific. Slight deviations from the typical value will have no consequences regarding system integrity (VGB).

***Limit Values***

Limit values are values that must be respected by the plant operators under any circumstances. Deviations include the possibility of materials corrosion and the consequences thereof. Immediate corrective measures are required.

***Control Values***

Control values are those parameters, which determine the entire situation of water chemistry regarding an optimum plant operation and thereby ensuring the absence of corrosion in the systems. They are selected because of their decisive importance and their exact measurability with state of the art analytical methods. For control parameters Action Levels are defined.

***Diagnostic Values***

Diagnostic values are values which supplement the entire picture of the applied water chemistry. Because diagnostic values are connected directly or indirectly with control values, they will enable to identify the root cause of deviations considerably.

***Action Levels***

Action levels are values which are defined for remedial actions to be taken when parameters are confirmed to be outside the control or diagnostic values. In such cases efforts should be made to bring the values within the appropriate limit within a certain time window. Such time limits can be taken from Table 2-3.

Table 2-3: Overview on Definitions and Time Windows

	<b>Abbreviations</b>	<b>Definitions/ Consequences EPRI</b>	<b>Definitions/ Consequences VGB</b>	<b>Definitions/ Consequences EdF</b>
Typical value	NV	No specification	Values resulting from undisturbed operation	Values resulting from undisturbed operation
Action Level 1	AL 1	7 days without power reduction	28 days without power reduction	7 days without power reduction
Action Level 2	AL 2	24 hrs	7 days	24 hrs
Action Level 3	AL 3	Immediate shut-down with temp. <250 °F	12 hrs before shut-down	Immediate shut-down

### 2.1.2 Review of Guidelines

Fruzzetti et al., 2004 reviewed the EPRI Chemistry Guideline and emphasised the desirability to go to a constant elevated  $\text{pH}_T$ , which may be at 7.1 and 7.3. The expected effect is a better control of crud generation especially at BOC and thereby the possibility to avoid AOA as well as high radiation field. As mentioned earlier, the EPRI Guidelines are updated every 4 years with the latest issue released in 2003. This revision 5 of 2003 consist out of two volumes which handle in Volume 1 the “steady state operation” and Volume 2 covers “start-up and shut-down chemistry.”

The authors are describing in their review the significant changes in the guidelines since Revision 4. Thereby they follow the various sections of Revision 5 which cover e.g. under Section 2 the technical basis for the water chemistry.

The first point they are mentioning is, that recent data and a revised statistical evaluation of test data were discussed regarding their impact on primary water stress corrosion cracking. This evaluation indicates that the use of higher Lithium levels, required for elevated  $\text{pH}_T$  regimes, results in little or no penalty in the characteristic time to PWSCC. Beside the test data the operation experience shows e.g. in French, Swedish and US plants, that the  $\text{pH}_T$  has little influence on PWSCC.

Another point is the discussion with respect to the effect of hydrogen on PWSCC which was revised on the basis of recent information. These data show that hydrogen concentration associated with a higher crack growth rate varies as a function of temperature.

The second point they are mentioning is that in Revision 5 of the Guideline an extended discussion was included on the benefits of constant high pH regimes with regard to crud, fuel deposit and radiation dose rate. This discussion is especially relevant to plants with high duty cores where risks of fuel deposits and the consequences thereof are a concern.

The third item in this Review is related to the use of Zinc in PWR plants. One aspect is the mitigation of PWSCC by Zinc and the conclusion that Zinc can stop the initiation of PWSCC but not the crack growth, Hickling, 2004. The second issue related to Zinc is the reduction of the shut-down dose rates. This chapter was updated in Revision 5 in order to reflect the continuous encouraging results from US and German plants

Some US plants, however, are reluctant for the time being to introduce Zinc because they are waiting for results from units with high duty cores, e.g. in Germany.

The fourth major item included in Revision 5 of the EPRI Guidelines is the influence of Primary Water Chemistry on corrosion on fuel cladding and on core performance. It emphasises the importance of crud to corrosion of cladding and the influence of crud on axial offset anomaly (AOA). According to Fruzzetti, the new EPRI Guidelines contain also a high duty core index (HDCI) which is calculated from the average heat flux of the peak fuel assembly, the cooling flow, and the coolant exit temperature of the plant.

Updates were also made on the effect of high silica coolant levels on fuel performance, indicating that with silica levels of up to 3 ppm no adverse effect could be observed.

The fifth point is a discussion on test results regarding low temperature crack propagation in thick parts made from nickel-base-alloys like X750, 82, 52 and 690, and how this cracking is affected by hydrogen levels in low temperature water.

In Section 3 of Revision 5 of the EPRI PWR Water Chemistry Guideline a quantitative chemistry guidance description is provided. It emphasises again the desirability of using a constant elevated  $\text{pH}_T$  between 7.1 and 7.3 for all plants. The guidance also reflects the two major concerns regarding high  $\text{pH}_T$  regimes, which are: (1) the effect of higher Lithium on PWSCC and (2) the effect of higher Lithium on fuel cladding corrosion. It was also agreed to raise the level at which consideration of a fuel vendor review is indicated as being appropriate from 2.2 to 3.5 ppm.

In the “Reactor Coolant System Power Operation Diagnostic Parameters”, Zinc was added as a parameter. This should be understood as a recommendation to all PWRs to inject Zn for dose reduction benefits.

Both, Section 2 and 3 of the EPRI Guidelines are considered to optimize the PWR Chemistry as shown in Figure 2-6.

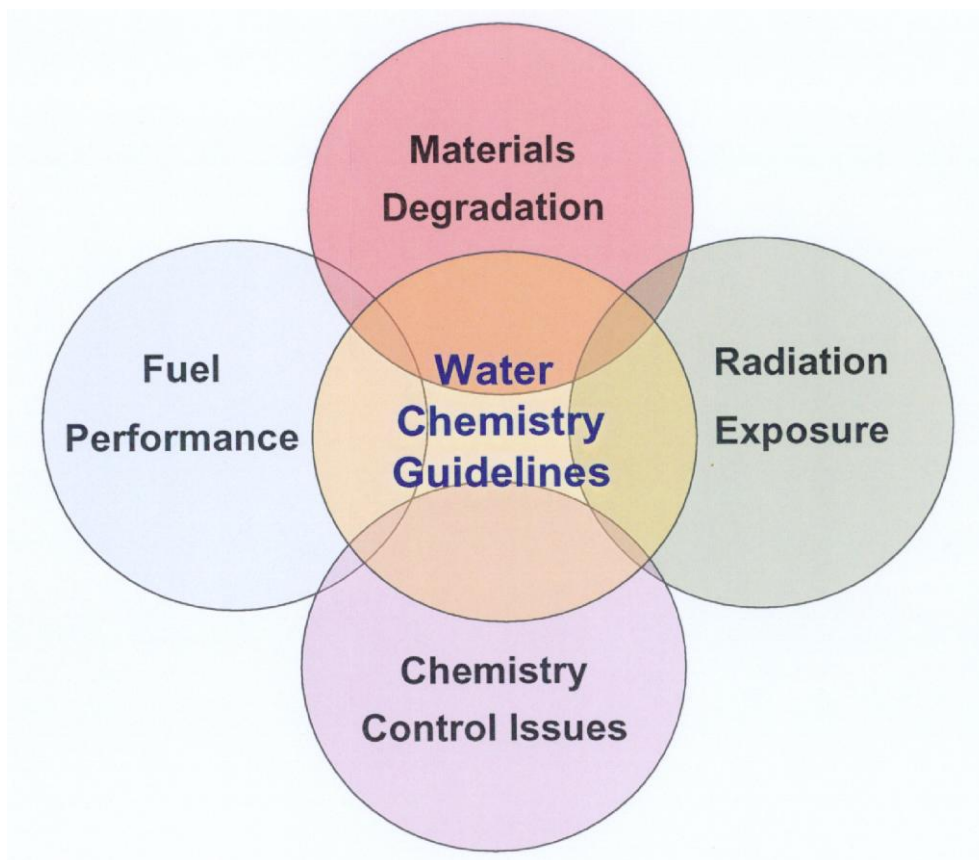


Figure 2-6: PWR Chemistry Optimization, Jones, 2004(a).

Fruzetti et al are listing numerous other changes in the Guidelines which are described in additional sections or appendices. These include:

- Section 4: Methodology for plant specific optimization
- Appendix A: Calculation of  $pHT_{T,}$  and Data Evaluation methodology
- Appendix B: Chemistry control of supporting Systems
- Appendix C: Status of Enriched Boric Acid (EBA) Application
- Appendix D: AOA and Ultrasonic Fuel Cleaning
- Appendix E: Oxygen and Hydrogen Behaviour in PWR Primary Circuits
- Appendix F: Sampling Considerations for Monitoring RCS Corrosion Products
- Appendix G: Reactor Coolant Radio-nuclides
- Appendix H: Definition of High Duty Core
- Guidance with respect to oxygen control in pressurizers. This guidance was expanded to cover control of oxygen during shutdowns and start-ups

Volume 2 of Revision 5 of the EPRI Guidelines is covering start-up and shutdown chemistry. Shutdown chemistry get its importance from the trend to shorten the refuelling outage. Chemistry can contribute considerably to minimize the time from breaker trip to reactor vessel head lift. Three other important issues are plant degassing procedures, forced oxidation results, and pressurizer oxygen control.

In an analogous way, the start-up chemistry should try to achieve power ascension in a minimum of time and meeting corrosion product transport goals that might impact core performance in the new cycle.

The significant changes made to Volume 2 of Revision 5 are as follows:

- Descriptions of the morphology and properties of the newly discovered fuel crud constituents Bonaccordite and Zirconium oxide were added to the technical basis for start-up and shutdown.
- Methods for monitoring and controlling hydrogen and oxygen in the pressurizer during shutdown and start-up.
- A discussion regarding the use of acidic reducing conditions during mid-cycle outages.

- Plant experience that showed strong benefits from using the maximum practical RCS cleanup flow during shutdown.
- Discussion on the needs and methods to maintain oxidizing conditions in the reactor water through flood-up for minimizing activity release during that operation.
- Oxygen control strategy in pressurizer
- Description regarding use or non-use of reactor coolant pumps during shutdown, including when adding hydrogen peroxide.
- Discussion regarding the benefits of using higher cross linked resins.

According to the authors the start-up and shutdown Tables in Section 3 and 4 constitute the quantitative chemistry guidance in Volume 2.

The changes made in these Tables reflect the experience gained since the last Revision, including the topics noted above. It is also recommended by the authors, that the nuclear industry must develop methods about Low Temperature Crack Propagation (LTCP) in Nickel-base alloys which depend on temperature and stress intensities.

In summary, the significant modifications to Volume 2 can be identified as:

- Significant enhancement to fundamental knowledge to the morphology and properties of fuel crud and its effect on shut-down chemistry strategy.
- Many changes to the start-up and shutdown tables were incorporated, substantially enhancing the quantitative chemistry guidance.
  - Reflect lessons learned from experience since previous revision.
  - Discussion of Low Temperature Crack Propagation (LTCP) in Nickel base alloys.
- Enhanced discussion of methods for monitoring and controlling hydrogen and oxygen in the pressurizer during shutdowns and start-ups.

Since Revision 5, Volume 1 and 2, consisting out of several hundred pages, were not available as a hard copy, during the evaluation of the open literature it is difficult to make a final judgement on the various items mentioned in Fruzzettis review. However, in discussions with customers beneficial quantitative values may have become disclosed and may be used in further discussions.



In contrast to the EPRI PWR Chemistry Guidelines, the DRAFT German Guidelines, to be issued by VGB, consists out of a dozen pages. The currently existing Guideline (Richtlinie “R 401” DWR) is a successor of a document named VGB R 401, issued in 1998. The entire Guideline is revised with the intent, to fix only those things which apparently have shown to be important for plant operation. This resulted among others in a stronger separation in control and diagnostic parameters.

The term normal operating value was still kept in the new Guideline, whereas terms like” Richtwert “and” Grenzwert” were replaced by the international term Action Level. This implies that the revised VGB Guideline includes also experiences from other countries like in the US and in France.

In the Preamble of this Guideline it is emphasised that this document is the joint basis for the operation of all PWR plants in the VGB domain. It shall be the frame, where Operation Manual and Chemistry Handbooks are in use. The task of these Handbooks (operational and chemistry) is to apply the Guideline to plant specific situations

The Preamble is followed by a description of the application area of the Guideline and some definitions, similar to those shown above.

The RCS description itself consists of:

- The technical background, ending with a recommendation to apply the “modified Li/B-Chemistry”.
- Comments to the Additives and Impurities in all operational modes.
- The Specifications as the “core-part” of the document (see below).
- Special modes of coolant treatment like: (1) Use of Enriched Boric Acid (EBA), (2) Zinc Injection, (3) Hydrogen peroxide injection during shutdown for refuelling-outage, and (4) Chemical control of radioactive iodine during shutdown.

**RCS Coolant Specifications of VGB*****Specifications of the RCS Coolant during start-up (>120°C)***

Control Parameter	Limit Value
Chloride (mg/kg)	< 0.2
Oxygen (mg/kg)	< 0.1 Heat-up >170°C only when oxygen concentration is < 0.1 mg/kg with falling trend

Diagnostic Parameter	Typical Value
B (mg/kg) / B-10 (AT-%)	Physical parameter
Lithium (mg/kg)	According the plant specific pH program
pH (at 25°C)	Depending on the Li and B concentration
Conductivity (µS/cm at 25°C)	

***Specifications of the RCS Coolant during Power Operation***

Control Parameter	Typical Value	Action Level 1	Action Level 2	Action Level 3
Lithium <sup>1)</sup> (mg/kg)	>0.2 <sup>2)</sup> <2.1 <sup>3)</sup>	3)	3)	3)
Hydrogen (mg/kg)	2 <4 <sup>4)</sup>	<2	<1 >4	<0.5 >5
Oxygen (mg/kg)	<0.005	<sup>5)</sup>	<sup>5)</sup>	<sup>5)</sup>
Chloride (mg/kg)	<0.01	>0.1	>0.2	>1.0
Sulphate (mg/kg)	<0.01	>0.1	>0.2	>1.0

<sup>1)</sup> Depending on the plant specific pH program

<sup>2)</sup> During the transition to stretch-out operation several days of operation below the normal operating value possible

<sup>3)</sup> Values to be coordinated with the fuel manufacturer

<sup>4)</sup> Optimal area of 2-3 mg/kg is recommended

<sup>5)</sup> When controlling the hydrogen specifications, the normal operating value will not be reached.

Diagnostic Parameter	Typical Value
Boron (mg/kg) / B-10 (At-%)	Physical Parameter
pH (at 25°C)	Depending on Li, B, (NH <sub>3</sub> )
Conductivity (μS/cm at 25°C)	Depending on Li, B, (NH <sub>3</sub> ) <sup>1)</sup>

<sup>1)</sup> Ammonia sources can be residual hydrazine used during start-up or radiolytic formation out of N<sub>2</sub> and H<sub>2</sub> coming from the volume control tank. Ammonia has a little influence on RCS pH.

### ***RCS Coolant Downstream HP Injection Pumps***

Control Parameter	Typical Value	Action Level 1	Action Level 2	Action Level 3
Oxygen (mg/kg)	<0.005/<0.05 <sup>1)</sup>	>0.01	>0.05	>0.1

1) Permissible during injection procedures

### ***RCS Coolant from boron recycling<sup>1)</sup> / Make-up Water***

Diagnostic Parameter	Typical Value
Conductivity (μS/cm at 25°C)	<1 (with almost Boron free replacement water)
Oxygen (mg/kg)	< 0.05

1) The condensate from the Boron recycling system.

### ***Fuel Pool Coolant***

Control Parameter	Typical Value
B (mg/kg) / B-10 At %	Determined by Reactor Physics

Diagnostic Parameter	Typical Value
Conductivity (μS/cm at 25°C)	<30
Chloride (mg/kg)	< 0.2
pH Value at 25°C	4.3 - 6

In summary, the VGB Specification is a very comprehensive document which has been no subject of many changes during the last 15 years. When comparing these Guidelines with what is known from the EPRI documents, one has to consider several differences between US plants and PWRs in the VGB domain. The following issues can be identified:

- VGB plants are using Alloy 800 as Steam Generator Tubing Material. In order to form protective oxide layers of the spinel-type 1/3 of divalent and 2/3 of trivalent elements are necessary. Such a condition is exactly provided by Alloy 800 with the following consequences.
  - More protective oxide layers than on Ni-base alloys
  - Lower metal release rates
  - Lower corrosion product concentration in the coolant
  - No AOA problems despite of High Duty cores
  - No Zr-alloy corrosion due to unacceptable Li – concentrations in deposited crud
  - In addition, the Hot Functional Test (HFT) is performed under slightly alkaline and reducing conditions contributing to low level radiation fields
- Alloy 800 is immune against PWSCC
- VGB PWRs are not following the trend towards extended fuel cycles

The **French Chemistry Guidelines** are published by Stutzmann, 1997, and they were discussed recently by Bretelle et al., 2004, and Nordmann, 2005. Their discussion is concentrating on the strategy of primary coolant pH, which can be characterized as the search for the best pH. EDF always tried to keep the balance between radiation build-up and AOA on one side (pH too low) and the risk of PWSCC and Zircaloy corrosion on the other side (pH too high).

This basic attitude can be demonstrated by looking to the pH evolution in EDF PWRs. The slight variations were introduced always in accordance with some re-definitions of the chemistry goals, see Table 2-4.

Table 2-4: pH Evolution in EDF PWR units.

Year / period	pH – Li	Goal
1977	$0.2 < \text{Li} < 2.2$	1
1978 – 80	pH > 6.9 $0.6 < \text{Li} < 2.2$	2
1980 – 97	pH 7.0 $0.6 < \text{Li} < 2.2$	3
Since 1997	pH 7.2- $0.4 < \text{Li} < 2.2$	4

**Goal:**

- 1) pH<sub>300°C</sub> sufficient to warrant the generalized corrosion and to limit the fuel cladding corrosion.
- 2) Dose rate consideration – To avoid fuel deposits.
- 3) Decreasing Boron-Lithium coordination with Lithium constant at the end of the cycle to avoid low pH at the end of cycle and easier operation conditions, Figure 2-7.
- 4) According to IAEA recommendation, code calculations, and past experience with pH of 7.0 compared to 7.2, (the international experience), EDF decided to select the pH<sub>300°C</sub> of 7.2 instead of pH<sub>300°C</sub> of 7.0 as the target pH (Zone 1, Figure 2-8). The previous target band of Lithium did not take into account minimization of the dose rates. The new target of Lithium value 0.1 mg/kg is associated with the reduction of dose rates to help the operators to keep as much a stable pH as they can do, Figure 2-7.

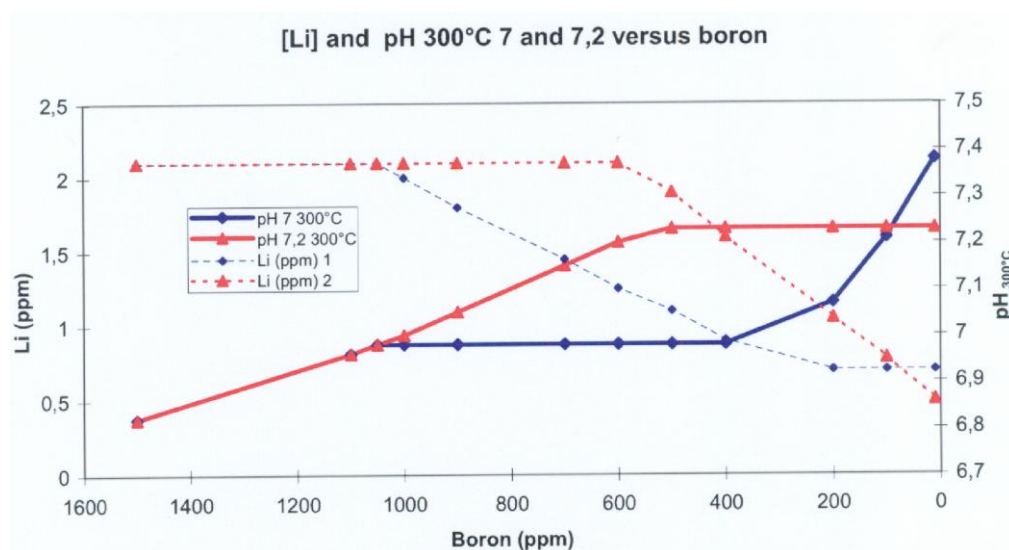


Figure 2-7: EDF Strategy regarding Li and pH<sub>300°C</sub> until now, Bretelle et al., 2004.

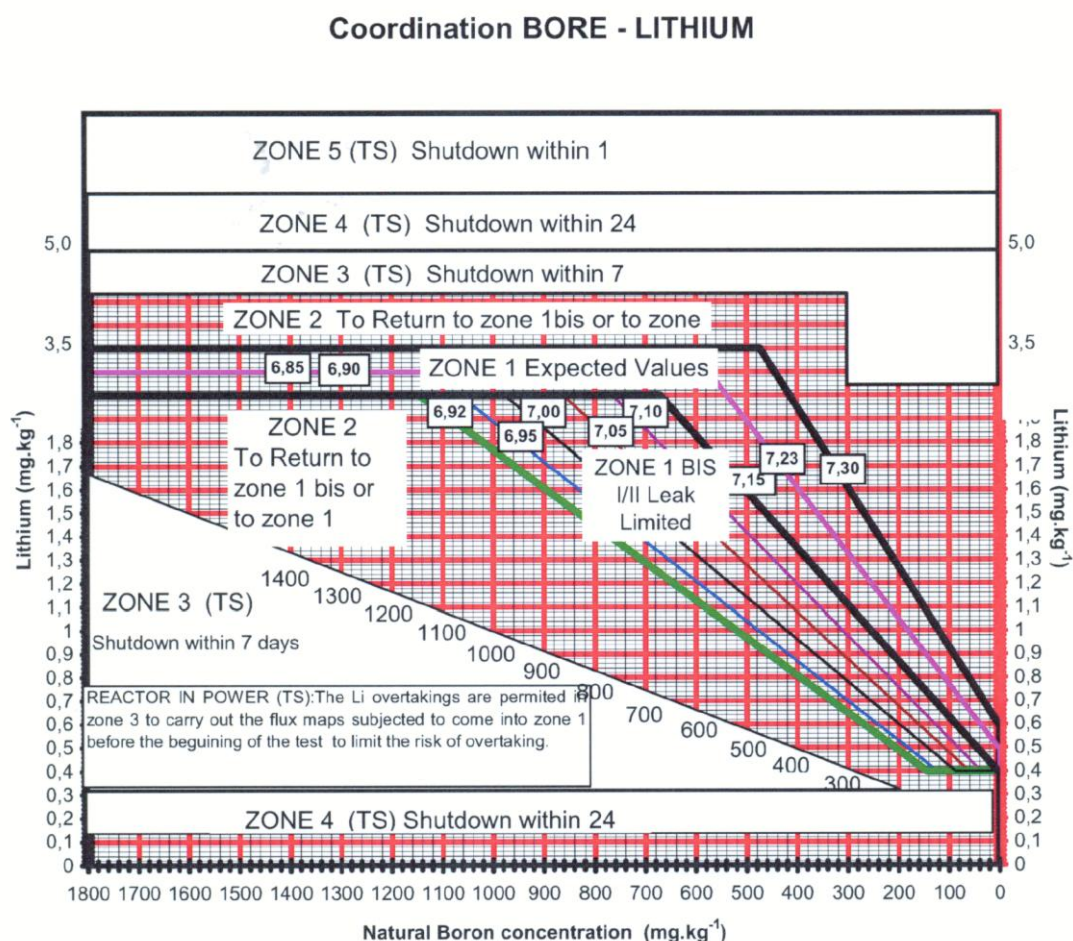


Figure 2-8: EdF Guideline for the coordinated B/Lithium Chemistry, Bretelle et al., 2004.

### **3 MATERIALS SELECTION FOR THE PRIMARY CIRCUIT (ALFRED STRASSER)**

#### **3.1 INTRODUCTION**

The objective of this Section is to identify the materials used in the major components of the primary coolant circuits of LWRs and the reason they were selected from the point of view of providing good corrosion resistance. Other criteria, such as mechanical properties, are considered peripherally. Fuel and core component materials are discussed in the ZIRAT reports.

The materials of construction for the primary circuit are originally selected during the design stage of the reactor, based on the knowledge available at the time, with most of these decisions dating from the 1960-ies and early „70ies. Originally these materials were considered to perform satisfactorily for the life of the reactor; however, for a variety of reasons the integrity of some of the components was impaired after many years of operation and they had to be replaced or repaired. A major reason for the need to replace the affected components has been corrosion, and specifically stress corrosion cracking (SCC) and irradiation assisted SCC (IASCC); general corrosion and erosion-corrosion have been some other significant issues that required component replacement. While these corrosion phenomena were well known at the time of the original reactor designs, several reasons accelerated their effects: unexpected impurity types and/or levels in the coolant, changes in water chemistry practices, excessive stresses induced in the materials during fabrication and/or installation, heat treatments that sensitized the materials, radiation effects on the materials and inappropriate material selection in the first place.

The decision to replace a large component, such as a steam generator, is a difficult one for the utility, since it is a very costly operation, not just due to the cost of the new component, but also the cost of the long shutdown time required for the replacement operation. However, in many cases it has to be done in order to maintain the required safety margins for operation and the cost has to be balanced against the permanent shut-down of the plant. Hence, the incentive to replace the initially selected materials with improved, longer lasting materials has spawned a large amount of research and development and changes in the materials used in the primary circuits. The most significant changes implemented and their causes will be discussed in this Section.

Prior to a detailed discussion of the primary circuit materials, the reader must realize that at the initiation of the LWR industry there were 8 nuclear steam supply system (NSSS) vendors of which each one made his own choice of materials. The PWRs were designed by Babcock & Wilcox (B&W), Combustion Engineering (CE), Westinghouse (W) and the original W licensees were KWU/Siemens, Framatome, and Mitsubishi. The licensees tended to follow the W designs to start with, but since they have become independent, are going their own way. The BWRs were designed by General Electric (GE) and Asea-Atom (AA) and the original GE licensees were AEG/KWU/Siemens and Hitachi/Toshiba. As in the case of the PWRs, the licensees, particularly KWU/Siemens, have gone their own way in design concepts as well as material selection.



Identification of all the materials used in every NSSS is a major project and clearly beyond the scope of this Section. Attention here will be paid to the major components and those that required repair or replacement due to water chemistry and corrosion issues.

## 3.2 PWRs

### 3.2.1 Overview of the Primary Circuit

#### 3.2.1.1 Components

A typical PWR reactor primary system is shown on Figure 3-1 and consists of the reactor vessel with its internals that support the fuel core, piping for the coolant recirculation system, a pressurizer, steam generators, pumps and valves. Auxiliary systems control the boron (B) and lithium (Li) additions and removals as well as a demineralizer, cleanup system for removal of impurities and a system to provide make-up water.

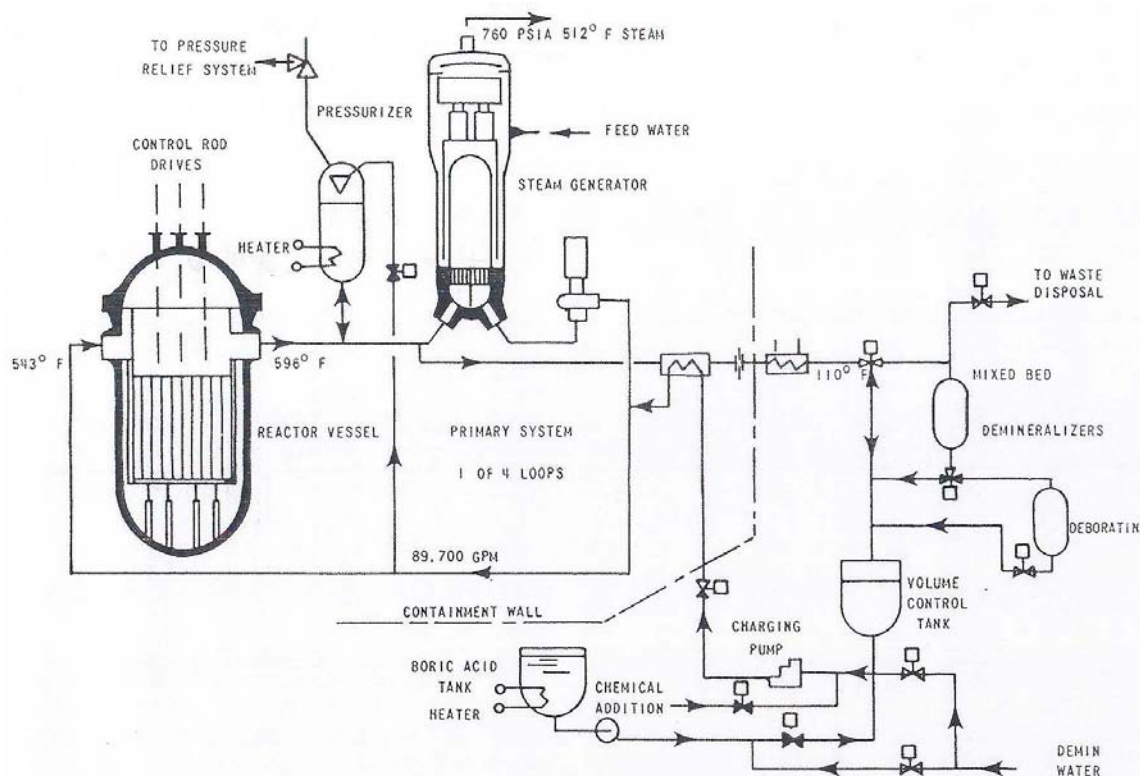


Figure 3-1: Primary Coolant Systems for a Large PWR, Cohen, 1985.

The primary system operates at a pressure of 2,250 psia (15.5MPa) and the *pressure boundary* consists of the reactor vessel, the recirculation piping, pressurizer, steam generator tubing and pumps as shown in Figure 3-2. There are *heat transfer surfaces* within the system that consist of the fuel cladding transferring heat to the primary coolant and the steam generator tubing transferring heat to the secondary coolant. The gamma heating of all the components within the reactor vessel is relatively minor, but is also removed by the coolant.



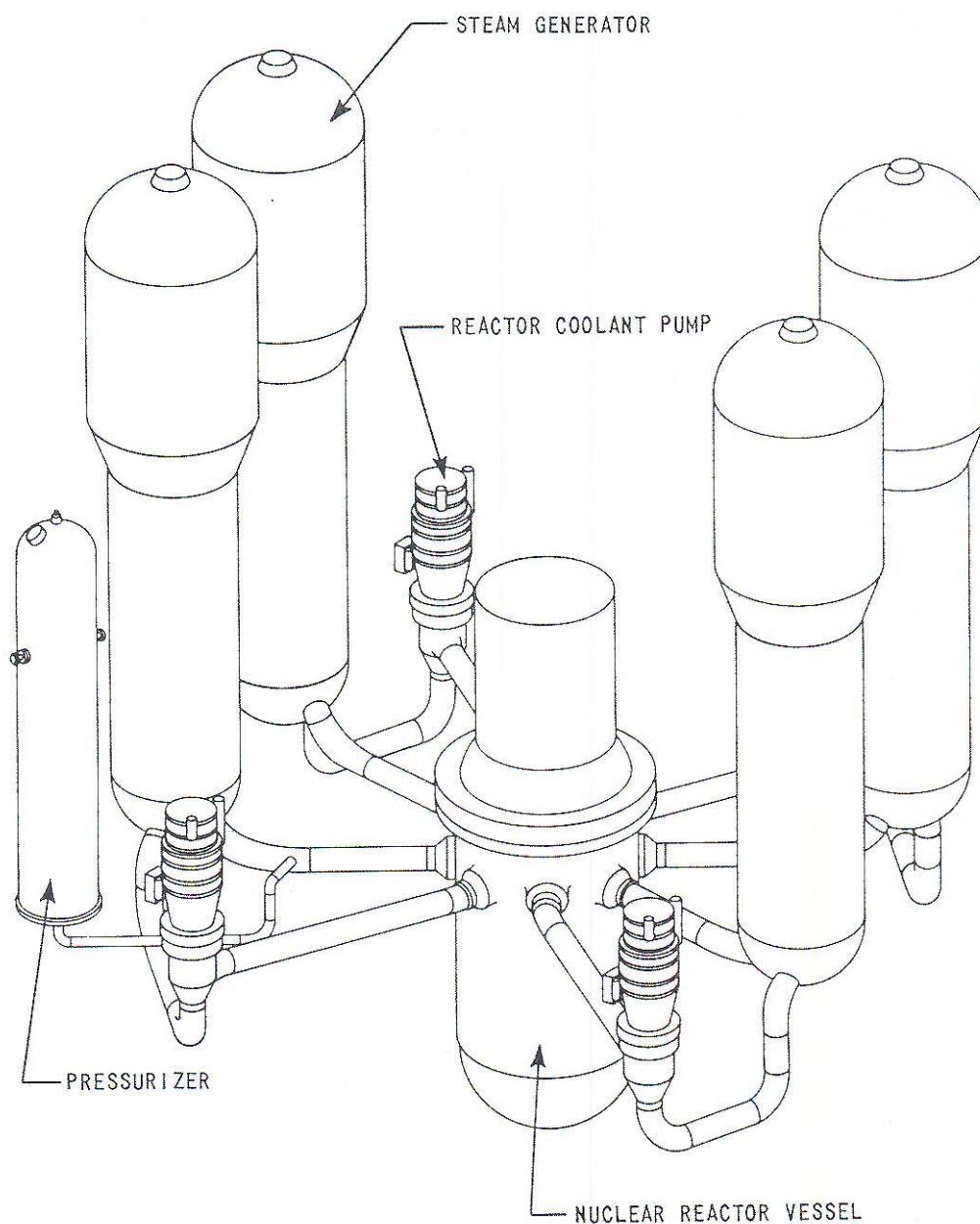


Figure 3-2: PWR Primary System – Pressure Boundary, Westinghouse, 2005.

The *structural components* within the reactor include the upper and lower support plates, the core barrel, the core support components and others shown on Figure 3-3. Structural components are used in the pressurizer and steam generator as well, but not all are in contact with the primary coolant.

## 4 PRIMARY CIRCUIT CORROSION

### 4.1 INTRODUCTION

Corrosion problems are the most costly issues today experienced by the operators of nuclear power plants. Examples for such issues are the stress corrosion cracking of stainless steel piping in boiling water reactors and the corrosion damage to steam generator tubes in pressurized water reactors. Chemistry based mitigation methods are identified and deployed wherever possible. However, where progress has been made in controlling the corrosion, minimizing its impact remains a continuing challenge.

Regarding the mitigation of corrosion problems in LWRs via chemistry changes, Jones, 2004(b), has reviewed the situation in the United States. He emphasised the economic importance of a safe and reliable operation of nuclear power plants. In addition, Nuclear energy also continues to set records by being the lowest cost, large scale expandable electricity source in the US. The average production cost is shown in Figure 4-1.

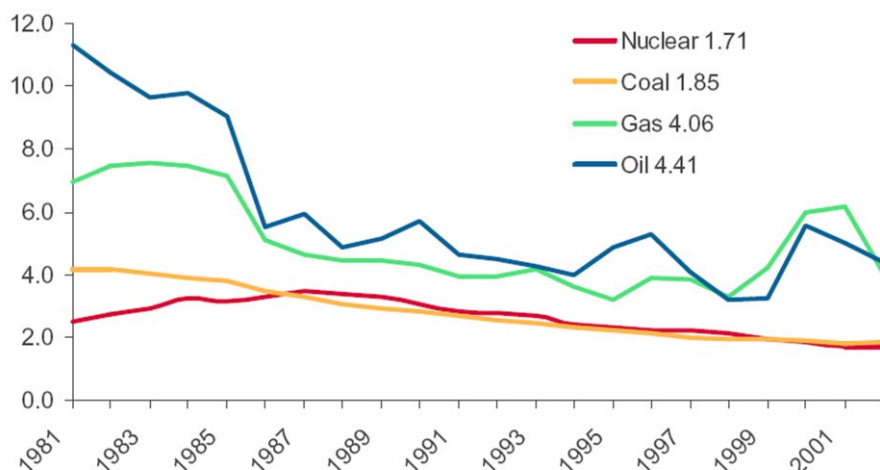


Figure 4-1: US Electricity Production Cost (1981 – 2002). In 2002 cents per kilowatt-hour, Jones, 2004(b).

As a result it is becoming apparent, that nuclear energy will be the central element in the portfolio for producing clean energy. The National Energy Policy, which was issued in May 2001, announced “Vision 2020”. This Vision 2020 means 50.000 MW of nuclear generating capacity shall be added to the grid by 2020. In order to get this a reality the capital cost of new nuclear plants must be reduced and the production cost of the existing must remain low. Materials integrity is an essential part to achieve the goals of Vision 2020.

However, historically the LWRs have suffered from various types of corrosion, including wastage, erosion corrosion, pitting, crevice and intergranular attack, stress corrosion cracking and corrosion fatigue. Especially, when corrosion cracking has affected pressure retaining components or core support structures, safety concerns were raised. Such corrosion problems impacting plant capacity especially peaked in the 1980s and the main issues were: Corrosion of reactor coolant system piping in BWRs (to be discussed later) and steam generator tubing corrosion in PWRs.

## 4.2 PRESSURIZED WATER REACTORS (ROLF RIESS)

### 4.2.1 Historical Perspective

Regarding the steam generator tube degradation problems, the following chronology can be given: Austenitic stainless steel was used initially for tubing in several early PWRs. As a result of corrosion problems such as caustic and chloride induced stress corrosion cracking, Alloy 600 was chosen. This was also stimulated by a decision of the Naval Reactors to use Alloy 600 in the Mill Annealed (MA) form instead of austenitic stainless steel. Table 4-1 summarizes this development and reflects also the chronology which led to the development of Alloy 800 and Alloy 690 TT (Thermally Treated). The same table indicates that the German OEM Original Equipment Manufacturer). decided already in the 1960s to switch to a modified Alloy 800 (800 NG (Nuclear Grade), followed by its first application in 1972. The basis for this decision was on one hand the SCC findings of Coriou with Alloy 600 and the early experience in large PWRs where numerous corrosion problems occurred. The most important diagram of those days is shown in Figure 4-2. The OEMs in France and Japan were Licensees of Westinghouse and they strictly followed the US decision regarding the SG(Steam Generator) -Tubing Selection.

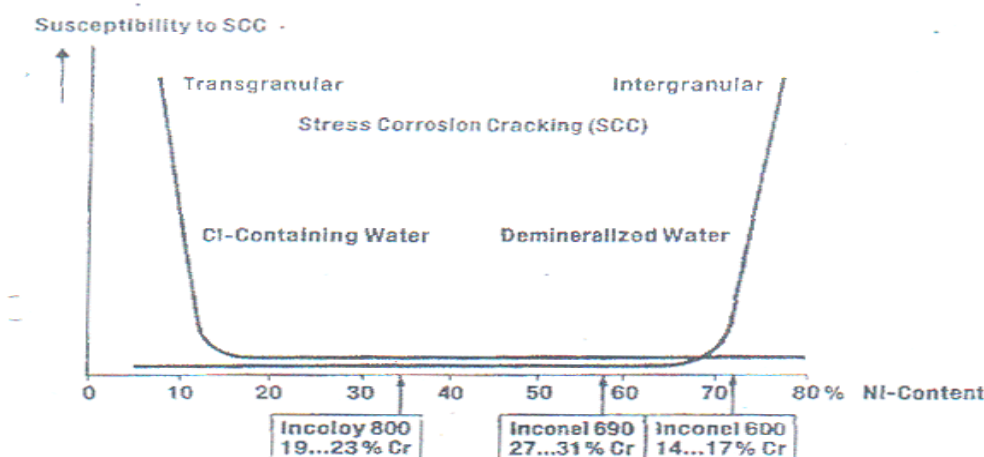


Figure 4-2: Stress Corrosion Cracking in 660°F (350°C) Water with 1000 ppm Chloride, Debray & Stieding, 1972

Table 4-1: PWR Steam Generator Design Evolution.

Model/Lead Plant (First Commercial Operation)	Typical Design T <sub>1</sub> (° C)	Hot Leg Heat Flux (Btu/hr/ft <sup>2</sup> )	Recirculating or Once Through	Tube Orientation	Tube Material*	Support Type	Support Material	Tube Sheet Expansion**	Flow Distribution Baffle /Y/N/	Preheater (Y/N)
<b>Government Laboratory Designs</b>										
Shippingport (1957)	283	29,000	Recirculating	Horizontal	SS then 600 MA	Hour-glass drilled holes	Carbon steel	PDR	Not applicable	N
N Reactor (1965)	279	67,000	Recirculating	Horizontal	SS then 600 MA	Drilled holes	Carbon steel	PDR	Not applicable	N
<b>Westinghouse and Licensees Designs</b>										
Early SS Designs (1961)	288 - 302	39,000	Recirculating	Vertical	304 SS	Drilled holes	Carbon steel	PDR	N	N
Early 600 MA Designs (1968)	310 - 318	43,000	Recirculating	Vertical	600 MA	Drilled holes	Carbon steel	PDR	N	N
Intermediate 600 MA Designs (1977)	317 - 322	44,000	Recirculating	Vertical	600 MA	Drilled holes	Carbon steel	FDE, FDR or FDR-KR	N	N
Later 600 MA Designs (1981)	323 - 324	48,000	Recirculating	Vertical	600 MA	Drilled holes	Carbon steel	FDR or FDR-KR <sup>1</sup>	Y	N
Preheater 600 MA Designs (1983)	326 - 330	49,000	Recirculating	Vertical	600 MA	Drilled holes	Carbon steel	FDR or FDR-KR	Y	Y
Model F Type (1983)	326	53,000	Recirculating	Vertical	600 TT	Broached holes	405 SS	FDH or FDR-KR <sup>1</sup>	Y	N
Preheater 600 TT Designs (1986)	326	49,000	Recirculating	Vertical	600 TT	Broached holes	405 SS	FDH or FDR-KR	Y	N
Latest Replacement Designs (1988)	See note	44,000	Recirculating	Vertical	690 TT	Broached holes	405 SS	FDH or FDR-KR <sup>1</sup>	Y	Y

## 5 DOSE RATE BUILDUP AND CONTROL

### 5.1 PRESSURIZED WATER REACTORS (ROLF RIESS)

The long term trends in occupational radiation exposure per plant and year are shown in Figure 1-1 for pressurized water reactors. This Figure is shown as courtesy of FANP GmbH and it is an update of Figures shown by Riess & Marchl, 2000. This Figure confirms that the Nuclear Industry has been successful in reducing radiation exposures of PWRs within the past decades. For the 4 countries shown, the average value for occupational radiation exposure approaches 1 – 1.5 Man-Sv per plant and year. These values, however, are still above the goal for the EPR, e.g. <0.5 Man-Sv per year. In this section the options shall be identified which may allow to achieve this goal.

In order to discuss the data of Figure 1-1 it has to be recognized, that in the 1980s the highest values for occupational exposure has been in the US. In a similar range the older Siemens plants were suffering from occupational exposures in the range of 5-6 Man-Sv per year. About half of this value did exist in the older Japanese PWRs, which started with about 3 Man-Sv per year in the early 80s. The best performance in the early 80s was shown by the French units. Within the last 2 ½ decades the French units improved, however, only by factor of about 2, whereas the plants in other countries had greater improvement factors.

Remarkable is the success of the Siemens pre-convoy and convoy units. In their 2 decades of operation they had an occupational exposure of <0.5 Man-Sv per year. In the same range as the Siemens pre-convoy and convoy units Sizewell B can be found, which is not shown in Figure 1-1. The reasons why Sizewell B and the Siemens pre-convoy and convoy units are so successful shall be explained later.

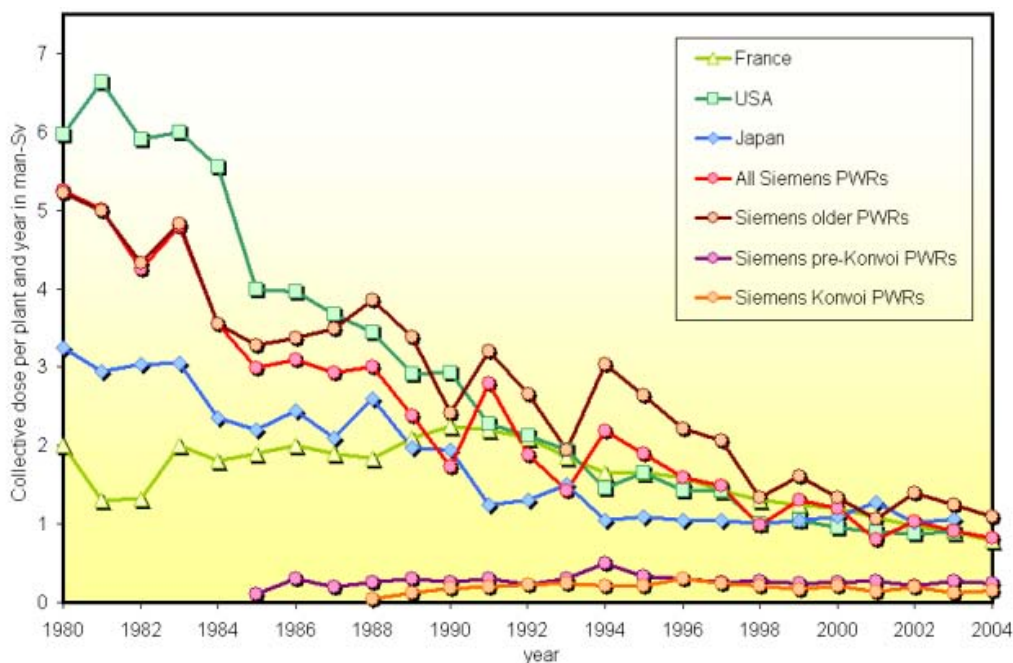


Figure 5-1: Occupational Exposure in PWRs: FANP 2005-08-01.

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### 5.1.1 Background

The primary source of PWR radiation fields is Cobalt 60, formed by a (n  $\gamma$ )-reaction from Cobalt-59 (elemental Cobalt). Cobalt-58 is formed by the Ni-58 (n-p)-reaction with fast neutrons and it is the second most important contributor to out-of-core radiation fields in older PWRs. However, in newer plants, where the major sources of Cobalt are removed, e.g. where the Cobalt-60 problem is minimized, the Cobalt-58 becomes the most important issue. In order to minimize the contribution of these radio-nuclides to out-of-core radiation fields, the sources of elemental cobalt and Nickel have to be identified and it has to be discussed how their contribution to radiation exposure can be controlled. The major source for elemental Cobalt (and thereby Cobalt-60) is the corrosion and wear of Cobalt base alloys (e.g. Stellites) and the Cobalt impurities in RCS structural materials (e.g. SG tubing). The major source for Nickel is the Steam Generator Tubing material. The radiation field build-up has then to distinguish between 3 major steps:

- Corrosion product release to the coolant, mainly from out-of-core surfaces
- To some extent: Deposition of this material on the fuel element surface and activation
- Re-dissolution of the activated products and re-deposition on out-of-core surfaces.

Each of these steps strongly depends on the primary coolant chemistry and on the chemical and physical properties of the elements cobalt and nickel under RCS conditions. Additionally, the first step also depends on the material properties, especially the SG tubing. However, the description of the Co- and Ni- (and thereby Co-60 and Co-58) behaviour in the RCS is complicated because of a variety of facts, like:

- The primary system is a non-isothermal system and the solubility and transport properties depend on the local chemistry conditions (Boron, Lithium and thereby the pH)
- Corrosion products in the coolant can be transported in three different forms:
  - as solid particles like metal or metal oxides
  - as colloids, e.g. in the form of hydroxyl complexes
  - as metal ions
- Corrosion products are metal oxides, e.g. Chromium forms under the reducing RCS conditions very stable Cr(III)-compounds with extremely low solubility.



- Deposits on the fuel surface consist primarily of non stoichiometric nickel ferrites to be formulated as  $\text{Ni}_x \text{Fe}_{3-x} \text{O}_4$ . In these nickel-ferrites the nickel can be replaced partially by cobalt in order to form complexes like  $\text{Co}_y \text{Ni}_x \text{Fe}_{3-x-y} \text{O}_4$ . Two facts can be derived from this situation:
- Chromium is not participating in the solution transport of the activity under normal operation.
- The contamination mechanism depends mainly on the solubility behaviour of nickel-ferrites.

In addition to nickel-ferrites, elemental nickel or NiO is found if not enough iron is present in order to form the ferrites.

- Regarding the chemical composition of the SG inner surface layer: It consists out of two layers:
- the inner layer is a nickel-chromium-iron spinel and
- the outer layer is more a nickel ferrite.
- Since the chromium spinels, the chromites are the most stable mixed oxides. They are the matrix to accumulate divalent radio nuclides like Co-58 and Co-60.

The overall process, e.g. the accumulation of the activity on the out-of-core surfaces is a complex matter of chemical and physical mechanisms interacting with each other. The various mechanisms and underline models shall be described later in this review. However, the conclusion is, that the coolant pH should be between 6.9 and 7.4 at 300°C in order to get a minimum in the radiation field build-up. This is also reflected in the primary coolant specifications which are set for 25°C and as they are described in detail in Section 2. Regarding the high temperature pH calculations, the OEMs (Original Equipment Manufacturers) are providing the necessary information to do the calculations.

In the following, each of the important factors shall be discussed in more detail including and emphasizing the plant experience gained in this field. As part of this discussion there will be a cross-check of available information with three possible and desirable objectives for an operating utility. Such objectives may be the following:

- The occupational radiation exposure shall not exceed 0.5 Man-Sv/year. Such value is the upper limit for the best performing PWRs which are Sizewell B and the Siemens pre-convoy and convoy units.
- The required annual outage time should be in the range of 16 days.
- Some radiochemical limits should also be fulfilled, which are
- Co-58 activity in the RCS coolant should be <5 GBq/t before RCS opening.
- Co-58 activity peak (spiking value) should be <16 GBq/t (or 0.43 Ci/t).

## 6 FUEL/WATER CHEMISTRY INTERACTION

### 6.1 PWR WATER CHEMISTRY (ROLF RIESS)

#### 6.1.1 Introduction

The primary coolant of PWRs, which contains boric acid (900-1800 ppm B at BOC) as a neutron absorber, is chemically conditioned by the addition of isotopically pure lithium (Li-7) hydroxide (2-5 ppm Li at BOC) as a non volatile alcalizing agent and of hydrogen.

Section 2 of this report contains a description of the Quality Requirements to be fulfilled by the RCS coolant, including a detailed explanation of the Lithium/Boron Chemistry modes.

Recently, an increasing number of PWRs is adding Zinc (5-40 ppb) in order to (1) reduce plant activation by reducing the metal release and by replacing Cobalt isotopes in the oxide layer and (2) minimize stress corrosion cracking of Inconel 600 material.

In VVER plants  $\text{NH}_3$  is added, which decays to  $\text{H}_2$  by radiolysis. Instead of LiOH, KOH is added, so that the pH-control is accomplished by the sum of the K- + Li- + Na-concentration (Li-7 is formed by the B-10 (n,  $\alpha$ ) Li -7). None of the VVER plants is adding Zinc like the PWRs.

From today's perspective it is most important to evaluate the factors, which are of greatest concern for the fuel element corrosion and which are the driving forces (problems) for Water Chemistry in the last 10 to 15 years.

These driving forces are moves to improve Plant Availability and Fuel Economics which can be characterized by:

- Changing to 18 and 24 months cycle
- Core up-rating
- Higher enrichment fuel, increased burn-up
- Low leakage cores combined with increased sub-cooled nucleate boiling

These moves - based on operational experience – caused concerns over coolant additives and impurities because the fuel elements in the operating plants (specially in the US) experienced heavy crud deposition at positions where sub-cooled boiling created two negative effects, namely (1) accelerated corrosion and (2) Axial Offset Anomaly (AOA).

The corrective actions considered to be effective, are:

- Higher pH Primary Water Chemistry
- Zinc Additions



For the pH strategy it is believed to avoid in any case  $\text{pH}_T$ -values of  $< 6.9$  by increasing the LiOH concentration above a long time valid value of 2-2.2 ppm Li. However, such Lithium increase may be a risk regarding the corrosion resistance of the Zirconium Alloys.

Specifically one environmental factor may be emphasized which is the corrosion product deposition on fuel surfaces, which can lead to increased cladding temperatures and increased corrosion rates. Such deposits have been identified as non-stoichiometric Ni-ferrites ( $\text{Ni}_x\text{Fe}_{3-x}\text{O}_4$ ), Ni-oxide or metallic Nickel. Such crud deposition is occurring specifically at positions with sub-cooled boiling and may cause accelerated corrosion defects locally and axial power shift by Boron precipitation (AOA).

Zinc addition may also lead to a more degrading crud at positions with high steaming rates. Thus, surveillance programs after introduction of Zinc are highly recommended, especially for PWRs with high duty cores. On the other hand, Zinc reduces the corrosion product release from system surfaces.

### **6.1.2 High pH Primary Water Chemistry**

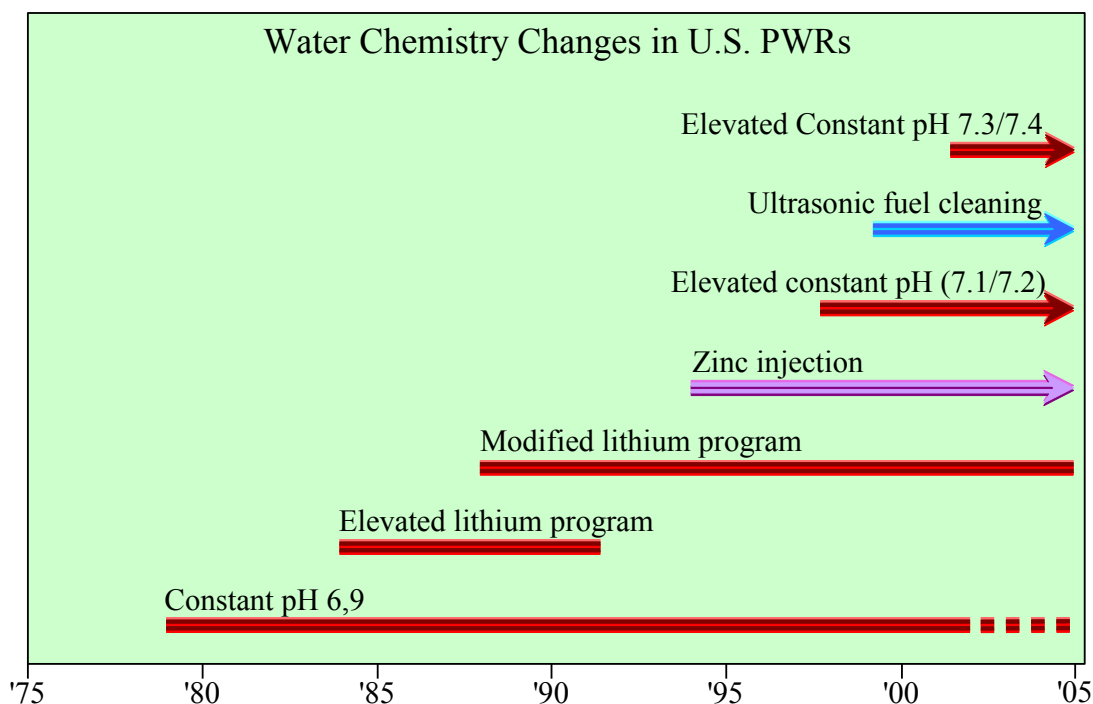
Regarding the application of all the Li/B-chemistries in operating PWRs there are major differences applied in the various regions and countries. Such variations depend also on the dates of the application. In the early 1970s worldwide the Lithium concentration was operated between upper and lower specified values. The first step to the coordinated chemistry was made in the late 70s/early 80s in order to reduce crud deposition on fuel rods and to minimize radiation fields. The selected  $\text{pH}_T$  was 6.9, which was also consistent with the  $\text{pH}_T$  for iron solubility minimum under PWR primary side conditions.

In the mid 1980s, many PWRs introduced the modified chemistry based on operational experience (see above), because at this time high and constant Lithium as a pH strategy produced better results than the coordinated chemistry, as far as crud deposition and radiation fields are concerned.

The German PWRs are still operating today with this type of chemistry and therefore the specifications, given by FANP GmbH and VGB respectively, have an upper Lithium limit of 2.1 ppm. Only in very specific cases, utilities can apply Lithium values higher than 2.1 with the consent of the fuel manufacturer.

In the second half of the 1980s in Ringhals an elevated Lithium strategy with an initial concentration of 3.5 ppm was tested. However, this project was stopped due to concerns for corrosion of Inconel 600 steam generator tubing and Zircaloy-4 cladding corrosion. Despite this kind of “Warning”, several US PWRs increased the cycle length and thereby the Boron concentration at BOC to values of  $\approx 1.500$  ppm Boron. The motivation for this step is described above. According to the general understanding the  $\text{pH}_T$  of  $>6.9$  can only be maintained when higher Lithium concentrations are allowed. This became operational practice by starting the cycle operation with 3.5 ppm Lithium, which means an elevated chemistry as described above.

An overview on the introduction of the various Li/B-strategies in the US can be seen in the paper of Turnage, 2004.



The latest developments in the US can be taken for example from Fruzzetti et al., 2004. In his review of the EPRI Water Chemistry Guideline he emphasized the desirability to go to a constant elevated  $pH_T$  which may be at 7.1 and 7.3 compared to a  $pH_T$  of  $< 6.9$  (modified Li/B –chemistry). The expected effect is a better control of crud generation especially at BOC and thereby the possibility to avoid AOA as well as high radiation field.

According to Fruzzetti, the new EPRI Guidelines contain also a High Duty Core Index (HDCI), which is calculated from the average heat flux of the peak fuel assembly, the cooling flow, and the coolant exit temperature of the plant.

A modified Lithium program was still not sufficient for plants with an extended fuel cycle and high duty cores because they still experienced AOA. This leads to the consideration, that an elevated coordinated chemistry may be a solution to the problem. The first test with this type of chemistry was performed in cycle 7 of Comanche Peak Steam Electric Station, Unit 2. Details of this test are described by Garzarolli, 2005, in Chapter 8 of ZIRAT 9. The most important message from this experiment is, that the oxide thickness was staying within acceptable limits, Figure 6-1.

## 7 CURRENT ISSUES AND MATERIAL DEVELOPMENT

As discussed in Chapter 4 the current corrosion issues are:

- SCC of Steam Generator Tubing
- PWSCC of Nickel base Alloys and weld metals
- SCC of Reactor Internals
- Fuel Cladding Integrity

Regarding these four items, progress must be made, because necessary repair or replacement work due to these phenomena is very expensive and enhances the production cost of electricity generated in nuclear facilities. Thus nuclear energy becomes less competitive.

The SCC of the steam generator tubing is a long term problem and deals mainly with Inconel 600 tubed steam generators. The specific problem from primary side is and has been the locations with higher residual stresses. Nowadays these locations are eliminated in new and replacement steam generators. An additional step forward in improving steam generator performance is the selection of Alloy 690 and Alloy 800 as current SG tubing material.

PWSCC of Nickel based Alloys and weld metal is the biggest challenge of the PWR Industry. Numerous investigations are ongoing to understand the very complex mechanism of this type of corrosion. However, the current mitigation solutions on the horizon seem to be Zinc injection and the optimization of the hydrogen concentration in the RCS.

In a reducing environment, core internals are less susceptible to SCC than under oxidizing conditions. However, under high fluences the behaviour of core internals is an area of uncertainty. Irradiation effects on toughness, IASCC, void swelling, and stress relaxation require more investigations and understanding. The NMCA method seems to be a very promising technology to create reducing conditions with a reasonable hydrogen injection, but there are several uncertainties that must be addressed. Of special concern is the crack flanking phenomenon, which probably demands on-line NM injection (OLNC). The OLNC technology must, however, be validated with more in-plant experience.

Fuel reliability is a permanent area of concern because fuel has been subject to design and duty changes. In addition, water chemistry was modified significantly in the last decade. Especially the use of NMCA together with zinc injection seems to increase the corrosion induced fuel failures in BWR plants. This change of water chemistry conditions puts a pressure on the utilities to further decrease the total inflow of corrosion products to the primary circuit.

Radiation field control is an area, where the PWR industry has been successful in the last decade by achieving in average an exposure rate of ~1 manSv per plant and year. However, activated corrosion products (especially Co-58 and Co-60) play an important role during shutdown.

In many cases they are released in concentrations, which require a time consuming cleanup process. Nowadays, where short outage periods are of top interest, the issue of minimum crud inventories gains a high priority.

Occupational exposures in BWRs have been significantly reduced during the last 20 years. Key factors have been Co reduction, improved condensate cleanup, development of efficient decontamination methods and the introduction of Zn injection. Further efforts to reduce Co sources are foreseen, as well as optimisation of the feedwater chemistry with respect to e.g. iron.

The subject of corrosion product generation and transport is also of vital interest in PWR plants for the fuel cladding integrity as well as for the phenomenon of AOA (Axial Offset Anomaly) or crud induced power shift (CIPS). With a small inventory of corrosion products both radiation field build-up during shutdown and the AOA problem could be controlled properly.

In the nutshell, the key area to mitigate these problems is the control of the crud source term. This leads to the question about our knowledge of this issue:

It is understood that there are oxide layers at the interface between water environment and metallic materials, playing a decisive role in the interaction between them. Water chemistry control is the control of the interfacial oxide layers from the water side. Nevertheless, the roles of both, the material and the water chemistry, in forming protective oxide layers, are not well understood, being sometimes phenomenological, qualitative, and even neglected. Deeper understanding is needed on formations, structures, properties, surface reactions etc. of the interfacial oxide films. Up to now, we only know beside an optimized water chemistry, that a properly performed hot functional test (HFT) and the addition of Zinc may have a beneficial effect.

Local chemistry inside cracks or crevices is sometimes largely different from bulk chemistry, but is very difficult to measure directly. Usually it is evaluated by simulation or model experiments. More information on crack chemistry is needed for the quantitative evaluation of crud propagation or some other local corrosion.

In situ monitoring techniques for example ECP monitoring or pH are very important but rather difficult especially in core. They are expected to work in combination with the proper sampling technology as key technology for preventive and predictive measures of plant aging and also to deepen the understanding of the phenomena of interest.

Finally it should be mentioned, that Water Chemistry Guidelines should be an instrument describing the water quality requirements necessary to achieve reliable operational results. Whether such Guidelines mainly shall be based on long term operational experience or on more short term test results, remain open.

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**APPENDIX A – UNIT CONVERSION**

<b>TEMPERATURE</b>		
$^{\circ}\text{C} + 273,15 = \text{K}$		
$^{\circ}\text{C} * 1,8 + 32 = ^{\circ}\text{F}$		
<b>T(K)</b>	<b>T (°C)</b>	<b>T(°F)</b>
273	<b>0</b>	32
289	16	61
298	25	77
373	<b>100</b>	212
473	<b>200</b>	392
573	<b>300</b>	572
633	360	680
673	<b>400</b>	752
773	<b>500</b>	932
783	510	950
793	520	968
823	550	1022
833	560	1040
873	<b>600</b>	1112
878	605	1121
893	620	1148
923	650	1202
973	<b>700</b>	1292
1023	750	1382
1053	780	1436
1073	<b>800</b>	1472
1136	863	1585
1143	870	1598
1173	<b>900</b>	1652
1273	<b>1000</b>	1832
1343	1070	1958
1478	1204	<b>2200</b>

<b>MASS</b>	
<b>kg</b>	<b>lbs</b>
0,454	<b>1</b>
<b>1</b>	2,20

<b>CONVERSION OF DIMENSIONS</b>	
1 Sv	= 100 Rem
1 Ci	= $3.7 \times 10^{10}$ Bq = 37 GBq
1 Bq	= $1 \text{ s}^{-1}$

<b>DISTANCE</b>	
<b>x (μm)</b>	<b>x (mils)</b>
0,6	0,02
<b>1</b>	0,04
5	0,20
<b>10</b>	0,39
20	0,79
25	0,98
25,4	<b>1,00</b>
<b>100</b>	3,94

<b>PRESSURE</b>		
<b>bar</b>	<b>MPa</b>	<b>psi</b>
<b>1</b>	0,1	14
10	<b>1</b>	142
70	7	995
70,4	7,04	<b>1000</b>
<b>100</b>	10	1421
130	13	1847
155	15,5	2203
704	70,4	<b>10000</b>
<b>1000</b>	100	14211

<b>STRESS INTENSITY FACTOR</b>	
<b>MPa√m</b>	<b>ksi√inch</b>
0,91	<b>1</b>
<b>1</b>	1,10