

# Water Chemistry in Boiling Water Reactors – A Leibstadt-Specific Overview

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**Abstract:** The boiling water reactor (BWR) consists of two main water circuits: the water-steam cycle and the main cooling water system. In the introduction, the goals and tasks of the BWR plant chemistry are described. The most important objectives are the prevention of system degradation by corrosion and the minimisation of radiation fields. Then a short description of the BWR operation principle, including the water steam cycle, the transport of various impurities by the steam, removing impurities from the condensate, the reactor water clean-up system, the balance of plant and the main cooling water system, is given. Subsequently, the focus is set on the water-steam cycle chemistry. In order to fulfil the somewhat contradictory requirements, the chemical parameters must be well balanced. This is achieved by the water chemistry control method called 'normal water chemistry'. Other additional methods are used for the solution to different problems. The 'zinc addition method' is applied to reduce high radiation levels around the recirculation loops. The 'hydrogen water chemistry method' and the 'noble metal chemical addition method' are used to protect the reactor core components and piping made of stainless steel against stress corrosion cracking. This phenomenon has been observed for about 40 years and is partly due to the strong oxidising conditions in the BWR water. Both mitigation methods are used by the majority of the BWR plants all over the world (including the two Swiss NPPs Mühleberg and Leibstadt).

**Keywords:** Boiling water reactor · BWR · Noble metal chemical addition · NMCA · Water chemistry · Zinc addition

## 1. Introduction

### 1.1. Main Goals of the BWR Water Chemistry

The main goals of the water chemistry of light water reactors are to provide a safe, undisturbed and environmentally compatible operation of the power plant. This means in particular: i) to minimize corrosion release and to prevent corrosion degradation of systems and components, ii) to minimise radiation fields and iii) to ensure compatibility with the fuel. These requirements are

fulfilled in boiling water reactors (BWR) by the 'classic' water chemistry control method called 'normal water chemistry' (NWC). NWC operation of BWR plants involves the use of pure water of low conductivity and low impurity content. The reactor water conditions are oxidising due to the presence of dissolved oxygen and hydrogen peroxide deriving from radiolysis in the core. As a result it has been observed for about 40 years that the oxidising conditions in BWR plants may lead to stress corrosion cracking (SCC) in components and piping made of stainless steel. The resistance to SCC can be increased by shifting the electrochemical potential to more negative values using injection of hydrogen into the feed water. This method is called 'hydrogen water chemistry' (HWC) and is being increasingly utilised in BWR plants. Unfortunately, two main negative effects are associated with HWC application: i) a significant increase of the main steam line radiation levels due to the formation of more volatile  $^{16}\text{N}$ -species in the reactor water and ii) an increase in shutdown radiation fields due to structural changes of the oxide layers formed on inner system surfaces under reducing conditions. To mitigate the latter effect, a further method has been developed adding trace amounts of zinc (depleted zinc oxide,

DZO). To avoid the increase in main steam line radiation levels an additional method involving doping of the oxide layers by a catalyst in conjunction with the reduction of the feed water hydrogen concentration is applied: the method is referred to as 'noble metal chemical addition' (NMCA) [1–6].

### 1.2. Tasks of the Power Plant Chemistry Organisation

In order to ensure the functionality of all materials and components within the water circuits, the power plant chemistry organisation is responsible for the supply of a sufficient quantity of deionised water in the desired quality. It is also in charge of the operation of the water treatment systems. Adverse trends in the different subsystems must be detected at an early stage by tracking all species contained therein. Frequently, chemistry provides the only means to quickly react to emerging problems. For this purpose a multiplicity of analytical measurements is regularly performed in the chemistry laboratory, for the most part on a daily basis. A series of on-line monitors provide in addition information on changing conductivity, activity, and radiation levels. A quick response to changes allows the timely implementation of adequate countermeasures.

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## 2. Principle of BWR Operation

### 2.1. The Water-Steam Cycle

From the point of view of a chemist, a nuclear power station is a huge water treatment plant. In a BWR such as the Leibstadt NPP (for technical data see Table 1) with a thermal power of 3600 MW, about 120 tons of water circulate per minute. The water acts both as a moderator and as a coolant. The steam is produced in the reactor pressure vessel (Fig. 1a). It drives the high and low pressure turbines to produce mechanical power. Subsequently it is condensed in the condenser. The fluid is purified in the condensate polishing system by ion exchange and mechanical filters. The purified condensate is then reheated to near reactor operating temperature and pumped back into the reactor vessel, where the circulation starts again (Fig. 1b). Thus, in a BWR plant, the reactor vessel, the turbines and the condenser are arranged in one single circuit, the so-called 'water-steam cycle'. This is the reason why, in contrast to a PWR plant, the turbine building is part of the radiation controlled area. The access to certain locations of the building is restricted or even not allowed due to elevated radiation levels (<sup>16</sup>N-radiation). Radioactive surface and air contamination is possible, particularly when contaminated systems and components are opened for maintenance and repairing. All materials, equipments, and persons must be checked for contamination according to the radiation protection standards, which also apply to the rest of the buildings. A BWR plant requires more effort to monitor the radiological situation and to enforce and check the compliance of radiation protection standards compared to a PWR. The minimisation of

Table 1. Selection of some technical data of the Leibstadt NPP

<b>Power Data</b>		<b>Turbine</b>	
Thermal power of reactor	3600 MW	Steam flow rate	1815 kg/s
Gross electrical output	1220 MW	Turbine generator speed	3000 rev/min
Total house load	55 MW	Condenser vacuum	0.130 bar
Net electrical output	1165 MW		
<b>Reactor Data</b>		<b>Generator</b>	
Core coolant flow rate	11'151 kg/s	Terminal rating	1318 MVA
Pressure at rated condition	73.1 bar	Terminal voltage	27 kV
Feed water temperature	222 °C	Cooling stator/rotor	H <sub>2</sub> O / H <sub>2</sub>
Main steam temperature	286 °C		
Main steam pressure	73.1 bar	<b>Main cooling water system</b>	
Steam flow	1'985 kg/s	Flow rate	~33'000 kg/s
Steam moisture content	< 0.1%	Number of pumps	4
		Flow rate per pump	8250 kg/s
<b>Core</b>		<b>Cooling tower</b>	
Total mass of uranium	113.5 t	Type	Natural draft, wet
Average specific uranium rating	31.7 kW/kg U	Height	144 m
Fuel element average enrichment (in <sup>235</sup> U)	4%	Max. diameter	119.2 m
Number of fuel elements	648	Loss of coolant (evaporation)	720 kg/s
Fuel material	UO <sub>2</sub>	Cooling water temperature difference	17.4 °C
Fuel element average weight of uranium	175 kg		
Fuel element length	4470 mm	<b>Reactor building</b>	
Fuel element weight	292 kg	Height	58.6 m
Fuel rod material	Zircaloy 2		

the radioactive contaminated waste is also a particular challenge.

In Table 2 some typical impurities of the reactor water are summarised. In the majority of cases they are extremely low (less than 1 µg/kg or 1 ppb).

### 2.2. The Transport of Impurities by the Steam

A part of the impurities in the reactor water is transported by the steam. These are

most of the fission gases resulting from fuel failures, traces of contaminants, a small fraction of the radioactive species resulting from activation of the coolant water, and water-borne corrosion products and impurities [7]. In addition, a small amount of non-volatile impurities is transferred within the steam by carryover of water droplets. Volatile impurities and non-condensable gases resulting from air in-leakage and radiation decomposition of the primary coolant are

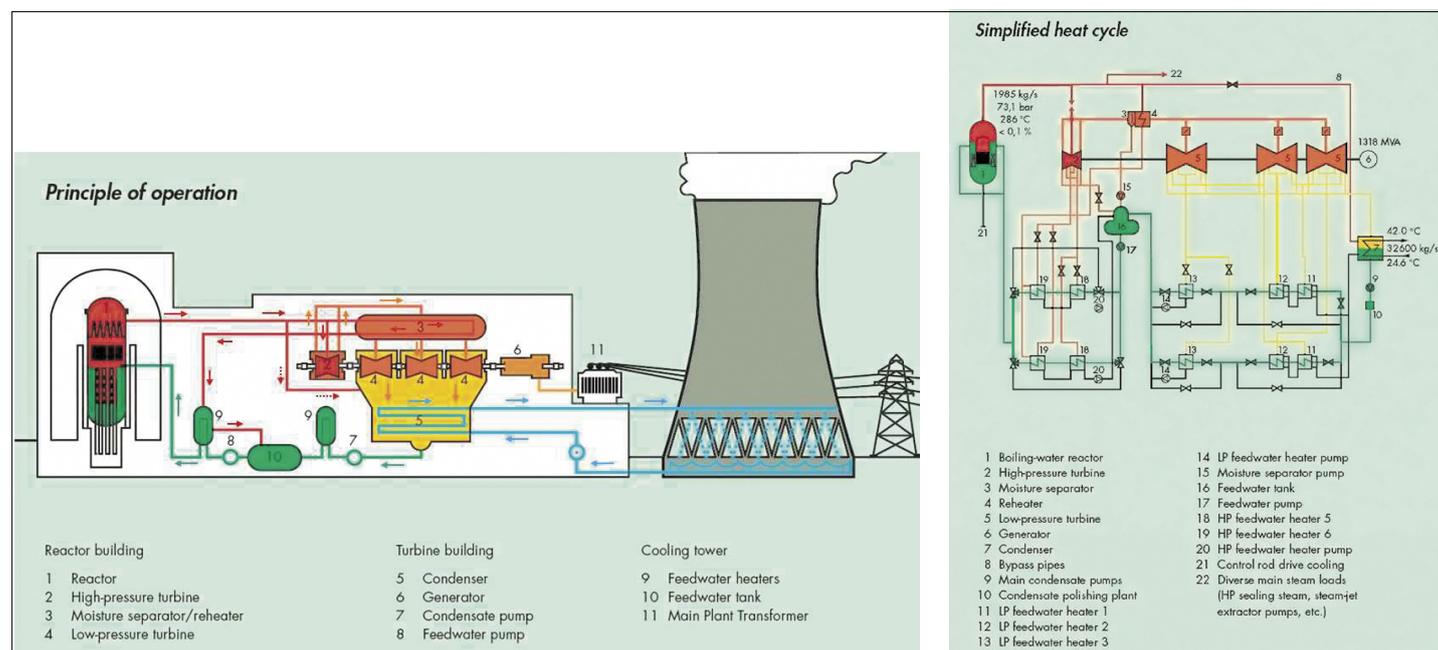


Fig. 1. a) Basic design of a BWR in terms of the two main water circuits: the water-steam cycle (red/orange = steam; green = condensate/feed water system) and the main cooling water system with cooling tower (blue). b) More detailed description of the water-steam cycle.

Table 2. Concentrations of the main species and isotope activities in the reactor water (average values)

Species	Concentration [µg/kg]	Isotope	Activity [Bq/m <sup>3</sup> ]
Cobalt	0.07	<sup>51</sup> Cr	1.4·10 <sup>+8</sup>
Nickel	0.15	<sup>54</sup> Mn	7.6·10 <sup>+5</sup>
Chromium	2	<sup>58</sup> Co	8.2·10 <sup>+6</sup>
Chloride	0.1	<sup>60</sup> Co	4.9·10 <sup>+6</sup>
Sulfate	0.8	<sup>65</sup> Zn	1.9·10 <sup>+6</sup>
Iron	0.4	<sup>131</sup> I	6.8·10 <sup>+6</sup>
Silicate	90	<sup>137</sup> Cs	3.9·10 <sup>+4</sup>
Oxygen	220	<sup>239</sup> Np	1.8E·10 <sup>+7</sup>
Zinc [added]	5	<b>Total activity</b>	1.1·10 <sup>+9</sup>
<b>Physical parameters</b>			
		Conductivity	0.08 µS/cm
		pH	7.1

evacuated from the condenser. Hydrogen in this gas stream is catalytically recombined with the oxygen to eliminate explosion hazards. The residual gas passes through large charcoal beds which provide a time delay for decay of most of the gaseous activity (the half lives of major volatile gases is short *e.g.* T/2 for <sup>16</sup>N is 7.1 sec, for <sup>137</sup>Xe is 3.8 min). The effluent of these beds is filtered through high efficiency particulate air filters (HEPA) in order to remove any traces of non-gaseous activities. The residual gas activity is measured and exhausted through the stack. The emission is balanced according to rigid safety standards and regulatory guidelines.

### 2.3. Removing Impurities from the Condensate

The condensate flow is treated by powdered ion exchange resin filters and/or by deep-bed ion exchangers. Deep-bed demineralisers are more effective to remove ionic impurities carried over in the steam or introduced by condenser cooling water leakage or by corrosion of condenser and steam system materials. Filter demineralisers are more efficient at removing particulates. Impurities exiting the condensate demineralisers are transported back to the reactor together with soluble and insoluble corrosion products released from the feed water system. These impurities are concentrated by the boiling process in the reactor vessel.

### 2.4. The Reactor Water Clean-up System

In order to maintain the reactor water quality at an acceptable level, the soluble

ionic impurities are removed by means of a reactor clean-up system, which can be a powdered resin filter demineraliser as well as a deep-bed demineraliser system. The clean-up flow generally amounts to 1% of the full power feed water flow. This means that the concentration of non-volatile ionic species in the reactor water will be about 100 times that of the feed water.

### 2.5. The Balance of Plant

Many auxiliary water-bearing systems represent the so-called balance of plant, including the fuel storage pool, the suppression pool, the water make-up system, various closed cooling water systems and the waste treatment systems. These systems are interrelated. The waste water is processed through the liquid radwaste system. Much of this water is recovered and recycled to condensate storage tanks. If necessary, a part of the inactive water is released to the environment in accordance with legislation. The radioactive residuals from the water treatment such as concentrates and sludges, as well as spent ion exchange resins are stored, worked up and finally conditioned. Different techniques are adopted for the solidification of these radioactive residuals; the one used at the Leibstadt NPP is immobilisation by means of a concrete matrix in a 200-liter drum. The resulting waste product fulfils the guidelines for the future final Swiss repository.

### 2.6. The Main Cooling Water System

The main cooling water circuit assures the condenser cooling (Fig. 1a). The water

is cooled down by partial evaporation in the cooling tower. The evaporated water has to be replaced with water from the river Rhine. The replacement water is decarbonated and conditioned prior to addition to the cooling circuit. A detailed discussion of the cooling water system chemistry is given by the paper 'Water Chemistry in Pressurised Water Reactors – A Gösgen-Specific Overview' in the present issue [8].

## 3. The Chemistry of the BWR Water-Steaming Cycle

In the introduction the main goals of the BWR chemistry and the methods of fulfilling its requirements were emphasised. The water chemistry influences the probability for the repair of piping and core components due to stress corrosion cracking. It governs the radiological conditions and therefore the radiation exposure of the plant staff both during operation and during outages. Finally, the water chemistry affects the corrosion durability of the fuel elements [9][10] as well as the amount of radioactive waste. However, these phenomena are to some extent contradictory. They need a balanced adjustment of the chemical parameters. Generally speaking, the highest degree of purity is required, in the reactor water as well as in the feed water. Impurities like cobalt, sulphate, chloride, and copper are detrimental and must be minimised. But other water ingredients like zinc, iron, hydrogen, and noble metals show beneficial effects. They are tolerated in some specified concentrations under balanced conditions. In fact, some of them are deliberately injected [2] (Table 3). The required concentrations are, as mentioned above, very small: they are in the range of 1 ppb (1 µg/kg). In contrast, an imbalance of the water content species can lead to unforeseeable detrimental consequences. Therefore changes in the chemical operation mode must always be well considered [11].

In the following, three selected topics are discussed in more detail: zinc addition, hydrogen injection and noble metal chemical addition.

### 3.1. Zinc Addition

Erosion and corrosion products deriving from structure materials of the secondary part of the water-steam cycle, *e.g.* from turbines, condenser, and pre-heater areas, are transported, essentially as insoluble particles, by the feed water into the reactor. Within the reactor vessel they are primarily deposited on the hot surfaces of the fuel elements. This has first of all a negative effect on the heat transfer properties of the fuel and can result in an enhanced corrosion of the cladding material along with a potential release of fission products into the reactor

Table 3: Overview of the substances in the reactor water, estimation of their properties and actions to be taken

Properties of some species in the reactor water		
Detrimental	Inoffensive	Beneficial
<b>Actions to be taken</b>		
Minimisation Removal	Admission Limitation	Addition Injection
Oxygen	Silicate	Zinc
Hydrogen peroxide	Nitrate	Hydrogen
Cobalt	Sodium	Platinum
Chloride		Rhodium
Sulphate	Iron	Iron
Copper	Boron	
Radioactive contaminants		

circuit. In addition, the elemental components of the corrosion layers are activated by neutrons. The radioactive nuclides can be re-mobilised into the coolant during operation transients and contaminate the wet surfaces of the primary circuit. This is the reason why in BWR plants with external recirculation loops maintenance work and periodical in-service inspections have to be performed mostly under challenging radiological conditions. Furthermore the activation products can be transferred by carry-over to the secondary part of the water-steam cycle. Again, this can lead to further contaminations of systems and components (fully volatile nuclides like the noble gases krypton and xenon feature a carry-over of 100%, non-volatile species such as sodium ( $^{24}\text{Na}$ ), cobalt ( $^{60}\text{Co}$ ) and caesium ( $^{137}\text{Cs}$ ) show a carry-over of about 0.001%, the partly volatile nuclides like iodine isotopes nearly 1%) [5].

One component of the deposited corrosion layers is the radionuclide  $^{60}\text{Co}$ . Due to its high  $\gamma$ -energy and relatively long half life of 5.3 a it is primarily responsible for the radiation fields in the reactor building. High collective doses can be accumulated in this part of the plant, particularly during outages and other maintenance work.  $^{60}\text{Co}$  is formed by neutron activation of the inactive isotope  $^{59}\text{Co}$ , which is a constituent of naturally occurring cobalt. The latter is found in several reactor structure materials made of austenitic steels and in other wear-resistant alloys. The experience in the Leibstadt NPP shows that the average annual  $^{60}\text{Co}$ -activity in the reactor water correlates very well with the mean dose rate measured at the recirculation loops during the main outage (Fig. 2). A high  $^{60}\text{Co}$ -activity in the reac-

tor water results in high radiation fields, in higher contamination levels and in elevated collective doses of the personnel.

Since the begin of operation of the Leibstadt NPP (1985), according to regulatory restraints, it was necessary to keep the mean dose rate of the recirculation loops below  $2 \text{ mSv h}^{-1}$ . Furthermore a guideline value for the power plant dose of  $4 \text{ Sv a}^{-1}$  was defined (today's guideline collective dose value is  $1.5 \text{ Sv a}^{-1}$  'only'). During the first years of operation the dose rate values increased rapidly beyond the limit. This fact could be attributed to the build up of the

$^{60}\text{Co}$ -containing corrosion layers. In 1990 it was decided to inject zinc in the form of zinc oxide with a natural isotopic ratio. The decision was taken according to observations made at that time, that power plants with (zinc containing) brass condensers showed substantially lower  $^{60}\text{Co}$ -contamination values and lower dose rates at the recirculation systems (due to the transfer of zinc from the condenser by the feed water into the reactor). However, three years later the addition of natural zinc was stopped again because of the unwanted neutron activation of  $^{64}\text{Zn}$  to  $^{65}\text{Zn}$  ( $^{65}\text{Zn}$  is a  $\gamma$ -emitter like  $^{60}\text{Co}$ , but with a short half life of 244 d) and replaced by the injection of depleted zinc oxide (DZO, depleted in  $^{64}\text{Zn}$ ) [12].

In Fig. 3, the runs of the  $^{60}\text{Co}$ - and  $^{58}\text{Co}$ -activities as well as the zinc concentration in the reactor water are plotted as function of operation years. The annual addition of zinc amounted to 8 to 20 kg. The step-by-step increase in the zinc concentration had to be reduced suddenly in 1996, after the appearance of fuel failures. Accelerated cladding corrosion near the spacers of the fuel bundles was attributed to a too low ratio of the trivalent iron to the divalent zinc and nickel ions in the reactor water. From that time on, the zinc addition rate was determined as a function of the iron slippage through the condensate polisher. The cobalt activities increased somewhat, reached a plateau later on and have decreased again during the past few years. In consequence the radiological conditions of the recirculation system and of the adjacent components have also improved (Fig. 2). The data demonstrate clearly the effectiveness of the zinc application, although the addition has to be

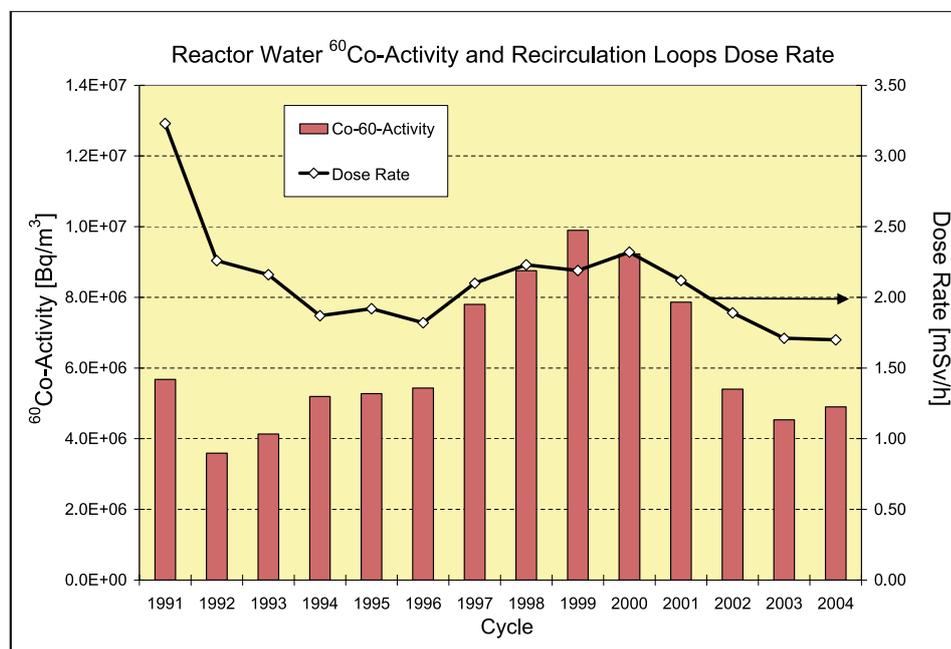


Fig. 2. Correlation of the annual average  $^{60}\text{Co}$ -activity in the reactor water with the mean dose rate at the recirculation piping (measured during the main outage)

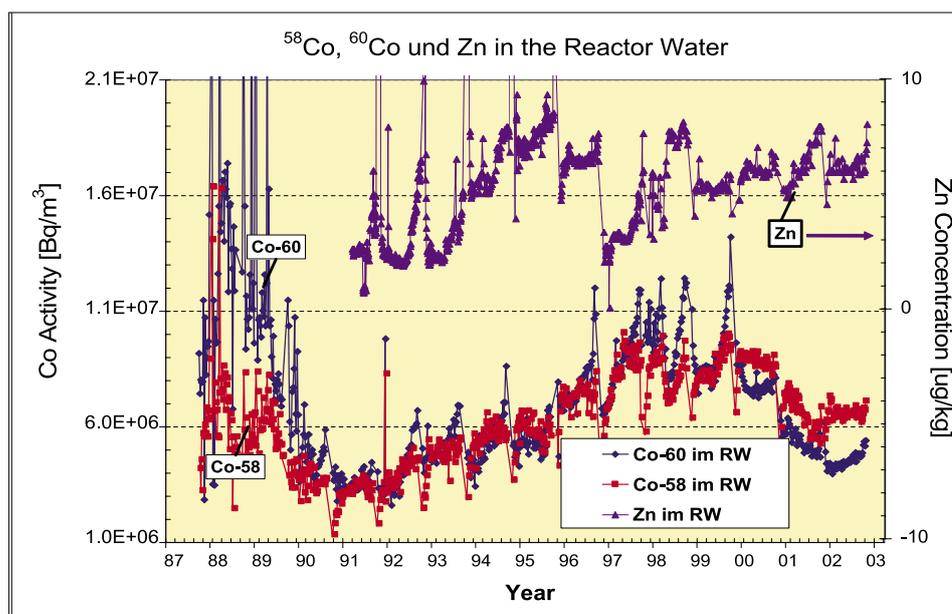


Fig. 3. Cobalt activities and zinc concentration in the reactor water during 16 years of operation

very carefully controlled, in order to comply with the fuel quality requirements. The current limit imposed on feed water zinc addition is defined according to the following equation:  $Fe/(Zn + Ni) \geq 2$ .

The beneficial effect of zinc addition is attributed to a reduction of the cobalt insertion into the oxide layers of the steel surfaces. Several mechanisms concerning the preferential uptake of zinc compared to that of cobalt were proposed. In [13] a narrowing of the pores of the oxide layers due to the presence of zinc and accordingly a decrease of the cobalt diffusion rate is suggested.

For the sake of completeness it should be mentioned that there are additional methods to reduce the cobalt build-up and deposition in the water circuits, *i.e.* the replacement of cobalt-containing components, the reduction of corrosion rates by the treatment of surfaces, and the use of electromagnetic filters.

Zinc injection has been implemented in 37 BWR plants all over the world (the first application was started in 1986). The Swiss BWRs Leibstadt and Mühleberg started in 1990 and 1998 respectively. Newly, zinc addition is being used in pressurised water reactors (PWR) too. The first purpose is, similar to BWRs, to reduce existing radiation fields and to optimise the radiation protection programmes, the second to mitigate stress corrosion cracking of nickel based alloys in primary cooling system.

### 3.2. Hydrogen Water Chemistry

Intergranular stress corrosion cracking (IGSCC) of stainless steel welds in boiling water reactors has been documented for more than 30 years. This phenomenon is well understood and proceeds at a slow and predictable rate [14][15]. For this reason the functionality of the concerned systems

is not questioned. The system integrity can be well monitored and appropriate countermeasures can be taken. However, the costs for inspections, analyses, repair, and the loss of power production are high. Therefore the BWR owner attempts to minimise the probability of IGSCC incidence. Even though no cracks are detected, it is more and more decided on the basis of external experience to implement prophylactic actions. There are three prerequisites for the occurrence of IGSCC: i) sensitive material properties of the weld heat affected zone, ii) the weld residual stresses and iii) the oxidising nature of the reactor water. For operating BWRs, the only practical method to reduce the risk of IGSCC events is to change the oxidising nature of the coolant.

The key parameter for the quantification of IGSCC in high purity water is the electrochemical corrosion potential (ECP) of the material. Under 'normal' conditions the ECP of BWR components is in the range of +100 and +300 mV (compared to the standard hydrogen electrode, SHE). In order to minimise the probability of commencing IGSCC or to mitigate the growth rate of already existing cracks, the ECP must be lowered to below -230 mV(SHE). Compare *e.g.* the PSI work of H.P. Seifert [16]. This can be done by hydrogen injection into the feed water – a concept called 'hydrogen water chemistry' (HWC). The hydrogen helps to recombine the radiolytically produced oxygen and to reduce hydrogen peroxide, the oxidants responsible for high ECP values. The oxidant concentrations are thereby decreased from values of several hundreds of ppb down to 10 ppb or less.

In order to protect the various core components against IGSCC, taking into account the different time and position dependent conditions, hydrogen must be injected into

the feed water at a minimum concentration of 2 ppm (mg/kg). Unfortunately, the positive impact of the hydrogen water chemistry to reduce the ECP is accompanied by an unwanted side effect, a multiple increase of the main steam dose rate, which is attributed to  $^{16}N$ - $\gamma$ -radiation. Under reducing conditions, the portion of volatile nitrogen species, which are transported by the steam to the turbines, increases. Therefore, at a feed water hydrogen concentration of 2 ppm, the radiation levels in the turbine building increase by a factor of about six. This fact causes considerable problems concerning radiation protection during inspection tours and work execution in the turbine building. In addition, problems related to the compliance with regulatory off-site radiation limits can occur. Consequently, further measures to reduce the radiation effects must be taken, *e.g.* the construction of additional massive shielding at selected components and on the building structure.

### 3.3. Noble Metal Chemical Addition

The main disadvantage of hydrogen injection, the increase of the main steam radiation levels, can be mitigated using another technique called 'noble metal chemical addition' (NMCA). This method minimises the negative side effect of hydrogen injection while still achieving the required ECP specification ( $\leq 230$  mV(SHE)). It is based on the fact that platinum group metals are electrolytical catalysts which help to recombine oxygen and hydrogen peroxide with hydrogen on the metal surfaces. They provide sites at which the mentioned species are dissociated and adsorbed, then readily undergo electron exchange reactions. The redox potential of the environment (Pt electrode) is governed by the redox recombination reaction of hydrogen and oxygen to water. Because of the very high exchange current density of the hydrogen oxidation reaction on Pt, the oxygen concentration drops to zero at stoichiometric concentrations of hydrogen in the bulk water and the redox-potential is close to the reversible equilibrium potential of the  $H_2/H_2O$  reaction. On pure stainless steel surfaces, the exchange current density of the hydrogen oxidation reaction is much lower. The corrosion potential of the stainless steels is lowered linearly with increasing hydrogen concentration, but the decrease to -230 mV needs much larger bulk water hydrogen concentrations.

The method of noble metal chemical addition was introduced by General Electric Company (referred to as NobleChem™) [3][17]. A plot of the corrosion potential variation on stainless steel in function of the added hydrogen concentration to the reactor feed water nicely shows the difference of the HWC and NMCA processes for lowering the corrosion potential to  $\leq -230$  mV (SHE) (Fig. 4).

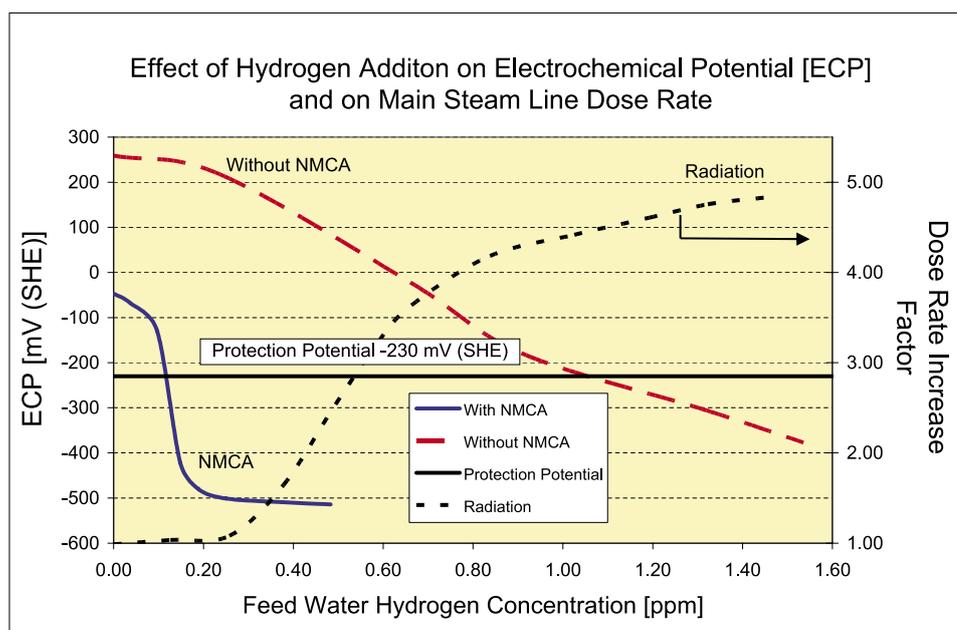


Fig. 4. Schematic plot of the dependence of the electrochemical corrosion potential (ECP) of stainless steel from hydrogen concentration in the feed water, with and without noble metal chemical addition. Also plotted is the dose rate increase factor due to the  $^{16}\text{N}$  activity in the steam.

#### 4. Outlook

So far hydrogen injection was implemented in 33 US and 8 European BWRs. The first NMCA application was made in 1996 at the Duane Arnold NPP in the USA. To date 25 BWR plants have adopted the NMCA technique: a majority in the USA, one BWR in Spain and one in Switzerland (NPP Mühleberg). However, the question if crack initiation could be prevented or crack growth rate decelerated under real power plant conditions, is not easily answered to this day. There are BWR plants without detected cracks using the mitigation methods. In other plants, where cracks were detected, it was not possible to show significant crack growth reduction up to now. More time is needed for NMCA-addition and to evaluate the experience. New developments such as the on-line NMCA technique will provide new results and additional experience [18]. However, in conjunction with NMCA applications several secondary negative impacts were also reported in some cases: increase of fuel cladding corrosion rates, increased deposition of corrosion products, mobilisation of activities and increasing radiation fields.

At the Leibstadt BWR core internals, no cracks were detected so far. Nevertheless, based on the worldwide experience it was decided to implement HWC. On the other hand, due to possible problems and uncertainties associated with the efficiency of the NMCA technique as reported above and taking into account the near-irreversibility of the noble metal addition, the application of NMCA is not intended. It is planned to start the hydrogen injection between 2006

and 2007. The construction of supplementary shielding on the turbine building and on selected components has already started. Numerous cameras are being installed in the future high radiation areas in order to reduce the number of inspection tours, and consequently, the collective dose of the personnel. In spite of the up to six-fold increase of the radiation level at the main steam lines, today's on-site and off-site radiation levels will not change thanks to the extensive additional shielding work.

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