

EFFORTS TO CONTROL RADIATION BUILD-UP IN RINGHALS

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ABSTRACT

It is well known that good control of the primary chemistry in a PWR is essential in order to minimise material problems and fuel damages. It has also been well established that the water chemistry has a great influence on accumulation of corrosion products on the fuel and the radiation build-up on primary system surfaces. Ringhals was one of the pioneers to increase operating pH in order to reduce radiation build-up and has now been operating for ten years with pH at 7.4 or (in later years) 7.2. Our experience is favourable and includes low radiation levels in the new (1989) steam generators of Ringhals 2. Ringhals 4 has operated almost its whole life at pH 7.2 or higher and it remains one of the cleanest PWRs of its vintage.

In addition to strict adherence to a stable operating chemistry, Ringhals is now working on a program with the aim to find optimum shut-down and start-up chemistry to reduce activity levels in the primary systems. A particular goal is to use the shut-down and start-up chemistry at the 1994 outage in Ringhals 3 in order to reduce doserates in preparation for the planned steam generator replacement in 1995.

The paper summarises the experience to date of the established operating chemistry, on-going tests with modified shut-down and start-up chemistry and other measures to limit or reduce the activity build-up.

INTRODUCTION

There are four nuclear sites in Sweden, and their 12 units provide about 50 % of the electricity in the country. The rest is mainly produced in hydroelectric plants. Of the nuclear plants, 9 are BWRs designed by ABB ATOM and 3 are PWRs designed by Westinghouse. The three PWRs are all located at the Ringhals site.

Ringhals is a four-unit site 60 km south of Gothenburg on the Swedish west coast. The site is owned and operated by Vattenfall.

Ringhals 1 is a 795 MWe BWR which has been in commercial operation since 1976. The plant was updated from 750 to 795 MWe in 1989.

Ringhals 2 is an 875 MWe PWR which has been in commercial operation since 1975. The steam generators were replaced in 1989 and the plant was updated from 800 to 875 MWe in 1990.

Ringhals 3 and 4 are identical PWRs with an installed capacity of 915 MWe each. Ringhals 3 started commercial operation in 1981 and Ringhals 4 in 1983. The steam generators of Ringhals 3 will be replaced in 1995 and at the same time modifications will be introduced to prepare for an increase in the capacity with 8 %.

ACTIVITY BUILD-UP AND MODIFICATIONS OF THE PWR PRIMARY CHEMISTRY

Chemistry During Operation

Activity build-up in Ringhals 2 was considered fairly normal during the first years after start up in 1975. The steam generator doserates, which are normally used to compare PWR activity levels, stabilised at 70 mSv/h (7 R/h) already after two years (1,2 EFPY) as can be seen in figure 1. Until 1979 the levels were about the same but we then saw a rapid increase in the channel head doserates. Another observation was very heavy crud layers on the fuel.

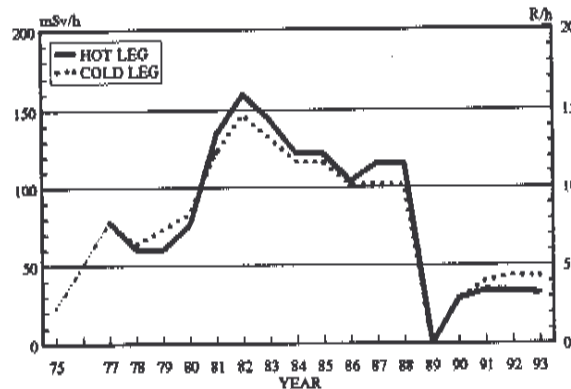


Figure 1. Average Doserates in the Steam Generator Channel Heads of Ringhals 2

Due to a considerable amount of work inside the steam generator channel heads we were quite concerned with the increasing doserates. Although we had been operating the plant within the Westinghouse chemistry specifications it was suggested that the causes of the problem were low Li and H₂ - concentrations. An increase in the minimum Li-concentration from 0.2 to 0.7 ppm was therefore introduced from the start-up after the 1979 outage and it was later verified that the rapid crud build-up on the fuel was halted.

Based on Westinghouse recommendations we made the next change, introduction of the original co-ordinated chemistry with a minimum Li-concentration of 0.7 ppm (the "dog leg" curve), after the 1980 outage. We now know that this chemistry will cause a transport of corrosion products from the system to the core, where it is activated, during the first part of the cycle. At low boron concentrations, and especially during coast down, the pH increases from 6.9 to as much as 7.4. This causes a change in the solubility of the corrosion products. This change in solubility has the effect that the activated corrosion products on the fuel will move to the coldest parts of the system, e.g. the steam generator cold leg side.

As seen in figure 1 our steam generator doserates continued to increase despite the chemistry modification. During one of our frequent mid-cycle shut downs late 1982 we found that the steam generator doserates had gone down from 135 mSv/h to 115 mSv/h during the few months of operation since the summer outage. Since we did not understand the reason for this nice break in the previously stable rise in steam generator doserates, we started to investigate possible causes.

During the spring of 1983 we came to the conclusion that the primary chemistry modifications at least had contributed to our high dose rates and this was confirmed by a paper¹ at the Bournemouth conference in the fall of 1983.

In order to improve the situation as fast as possible, we increased the pH from 6.9 to 7.1 after mid-cycle shut-downs in Ringhals 2 and 4 in late 1983. The pH was then kept constant for the rest of the cycle and ignoring the minimum specification for lithium. Ringhals 3 did not have a mid cycle shut-down and since we did not want to make the changes in Li (and pH) during operation we operated Ringhals 3 with constant pH of 6.9 through a continuous decrease in Li-content all the way to 0 ppm B and 0.35 ppm Li.

After the 1984 outages we wanted to operate all three units with a positive temperature coefficient throughout the cycle. This meant that we had to raise the maximum Li-content to 3.5 ppm for a limited time (25 to 106 days) in the beginning of the cycle. This level was above the fuel vendor specifications and also above the requirement in the technical specifications. We got approval for a one year test at a maximum level of 3.5 ppm Li in Ringhals 3 and 4 by our plant Safety Review Board, the authorities and the fuel vendors. Ringhals 2 was not operated with 3.5 ppm Li until 1985 because of fear for accelerated corrosion of the heavily crudded fuel. The permission from all parties did prescribe that a prolongation could only be accepted if extensive fuel examination could prove that this type of operation was not deleterious. The Li-levels were kept constant at 3.5 (2.2) ppm until we reached the pH of 7.4. From that point on the pH was kept constant until the end of the cycle. After this test, and after each of the following years with operation at elevated Li-levels, we performed fuel investigations. These included visual inspection and oxide thickness measurements every year and crud sampling some years. The results were encouraging, the fuel performed very well.

After the steam generator replacement in Ringhals 2 in 1989, it was uprated to 109% of the nominal capacity. After operation at about 80 % power for a couple of years, to save the steam generators, our fuel specialists and the fuel vendor feared accelerated fuel corrosion in one batch of fuel with cladding which was especially susceptible to corrosion. We therefore made an agreement with the fuel vendor to operate with maximum 2.5 ppm Li for a few weeks and then keep the pH constant at 7.25 for the remaining cycle. Our ambition was to keep pH as constant as possible in order to avoid solubility changes rather than aim for pH 7.4, which was thought to be the optimum pH.

The same year we also limited the maximum Li-concentration in Ringhals 3 and 4. The reason for this step was the preliminary result from on-going tests at Studsvik, which indicated shorter initiation time for stress corrosion cracking (PWSCC) in Inconel 600 steam generator tubing at elevated Li-concentrations. The Li-concentration was therefore limited to 2.2 ppm at these units.

In 1990 we discovered severe hydriding in the Ringhals 2 control rod guide tubes. It was later found that this hydriding was caused by the manufacturing process but initially the fuel vendor claimed that our chemistry could be the cause. Therefore the Li-concentration was limited to 2.2 ppm during the cycle 1990/91. The changes in the chemistry of Ringhals 2 is summarised in figure 2 and 5. What is not seen in these diagrams is that from our first modifications in the chemistry we tried to keep the conditions as stable as possible since we believe that chemical transients can be almost as bad for the radiation levels as a low pH. The Li-values used, and the resulting pH-values, are illustrated in figures 3 and 4 for Ringhals 3 and 4 respectively.

Shut down chemistry

At Ringhals we will use a shutdown procedure closely following the intentions of the EPRI guidelines. However, we will use the hot acidic reducing chemistry only during the cool down stage. The residual hydrogen is removed by slow controlled injection of hydrogen peroxide. Then we will add the main amount of hydrogen peroxide and use the time available to remove the dissolved activity during RCP operation.

Start up chemistry

The traditional start-up chemistry involved among other things, chemical degassing by the abrupt addition of hydrazine. Today we use slow controlled injection, which seems to allow a better removal of nickel. Lithium is

added when the Residual Heat Removal system (RHR) is disconnected and hydrogen is added just prior to dilution for criticality.

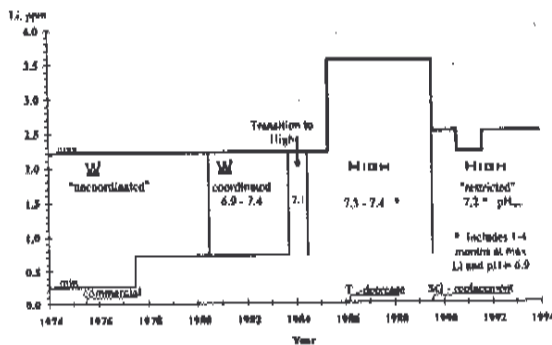


Figure 2. Ringhals 2 RCS lithium - pH history

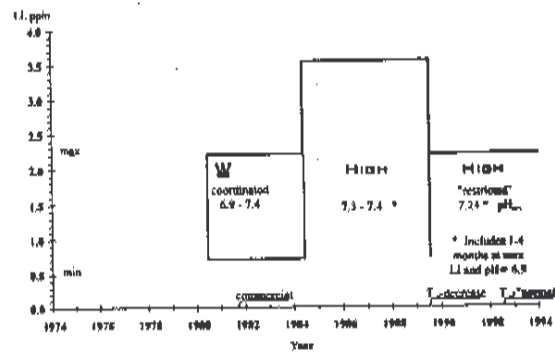


Figure 3. Ringhals 3 RCS lithium - pH history

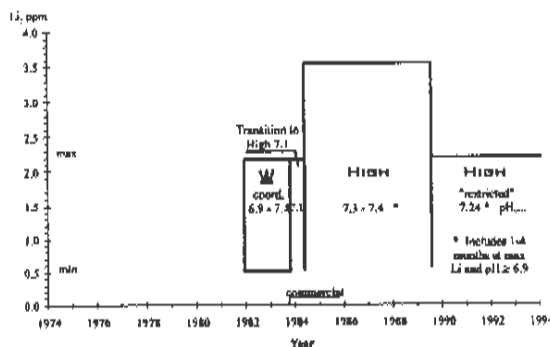


Figure 4. Ringhals 4 RCS lithium - pH history

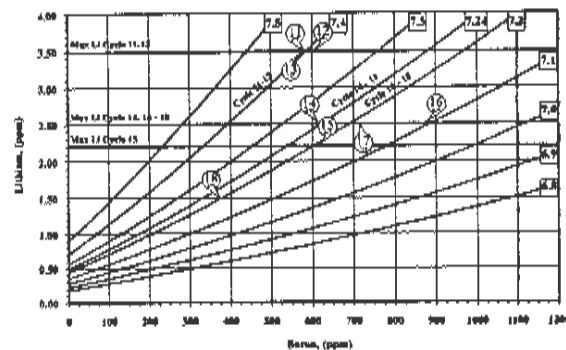


Figure 5. Ringhals 2. Diagram showing boron/lithium correlation at different pH. Figures in circles indicate boron concentration at the beginning of each cycle.

Results of the modified chemistry

Doserate Measurements in the Steam Generators

From 1983 onwards we have seen an almost steady decrease in Ringhals 2 steam generator dose rates as seen in figure 1, and in Ringhals 3 and 4 the activity build-up was more favourable than in Ringhals 2. As a matter of fact Ringhals 4, where we introduced the high pH chemistry after only 0.5 EFY of operation, is still the cleanest Westinghouse PWR. The early high build-up rate of Ringhals 2 and later improvements might also have been influenced by other effects. We know that some reactor vendors, including W who supplied our initial fuel, had problem with high Co-content in the Ni plating of the fuel spacers at the end of the seventies. Some European plants have experienced much higher dose rates than we did because of this. Later on all fuel vendors have got good control of the Ni-plating. They have also changed from Inconel to Zircalloy, which contains much less Co, in their fuel spacers and this change has reduced the potential for release of Co 60 into the Reactor Coolant System.

Dose Rate Measurement on the Primary System

The result of the measurements in the EPRI standard program indicates a continuous downward trend in dose rates on reactor coolant system. An example from Ringhals 2 is seen in figure 6.

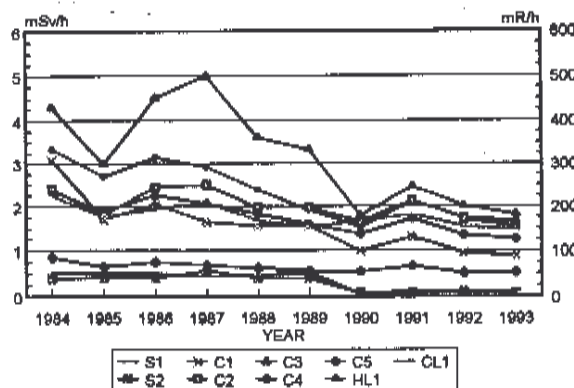


Figure 6. EPRI standard measurement program.
Dose rates on Ringhals 2 primary coolant system loop No 1.

Loose Contamination in Primary System Components

After the first cycle with high pH chemistry in Ringhals 2 we discovered that the amount of loose crud on the primary system surfaces was strongly reduced. This situation has been further improved after the changes in the shut-down chemistry (see below).

Activity Build-up in Regenerative Heat Exchanger after Decontamination

A positive long term effect of a decontamination is shown in figure 7. The regenerative heat exchanger in the Chemical Control (CS)-system was decontaminated in 1986 by the ODP-method. Although the dose rates have increased over the seven years since the decontamination was performed, they are still some three times lower than the original values. The question is, whether the ozone decontamination makes the oxide layer less prone to accumulate cobalt activity or if the new level just reflects a lower source term of Co-60.

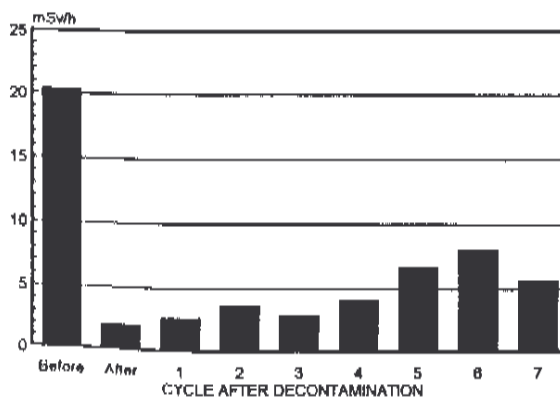


Figure 7. Dose rates on Ringhals 2 regenerative heat exchanger before and after decontamination. Average of 6 measurements.

Nuclide Specific Measurements on the Primary Circuit

One can note that the surface activity levels on the old system parts basically are of the same values as before the steam generator replacement.

The parts that were decontaminated with electropolishing during the steam generator replacement still have surface activity levels which are a factor of four, for Co 60, below the undecontaminated system surfaces.

The "elbows" that were replaced, were electropolished. The activity build-up to present level occurred during the first cycle but the surface activity levels of Co 60 still remain a factor of 14 below the undecontaminated system surfaces.

What happened when we reduced the pH from 7.4 to 7.2 or 7.24?

The change from pH 7.4 to 7.2 or 7.24 (from cycle 14 in Ringhals 2 and cycle 7 in Ringhals 3 and 4) does not seem to have a major influence on the radiation levels, but we are still convinced that pH 7.4 is closer to the optimum. What we have seen in all three units is a shift from having the highest doserates in the hot leg with pH 7.4 to somewhat higher doserates in the cold leg (figure 1). The high dose rate in the hot leg is probably due to residual activity from operation at "uncoordinated" and coordinated chemistry at pH 6.9.

Release of Corrosion Products at Shut Down

In recent years much attention has been devoted to shut down chemistry. Traditionally, we have always been using hydrogen peroxide addition as a normal part in the shutdown procedure. It ensures a concentrated release of activated corrosion products, and the subsequent collection on demineraliser, before opening up the systems for refuelling. The activity released during this oxidising phase originates solely from the fuel crud. Thus, it yields no reduction in the doserates from the system surfaces. The long term advantage is that it reduces the amount of material on the fuel, which may be activated in the next fuel cycle.

To reduce the dose rates from the system surfaces, the concept "Hot, Acid, Reducing Chemistry (HARC)" has been tested in many plants in recent years. The most striking results were obtained at Zion 2, where steam generator dose rates were reduced to about 40%. However, the total time used for this effort was some 70 days, which normally cannot be included in a shut down procedure.

We used HARC during the 1993 shut down of Ringhals 2 and our observations are in agreement with the Zion 2 data. However, the release rate during the reducing phase of the shutdown is much lower than during the oxidising phase after the addition of hydrogen peroxide. Although we did not observe any significant influence on the dose rates in the steam generators we found our channel heads extremely clean with regard to loose contamination.

We also noticed a small reduction in the Co-58 activity on the cross leg (some 20%) by on-line gamma scan and doserate measurements. The Co-60 activity was not affected. During the subsequent oxidising phase, a recontamination of Co-58 occurred, so only about half of the initial reduction in Co-58 persisted. There were also some indications that the doserates were higher in the loop where the RCP was stopped first, at the higher concentration of Co-58. It seems as if this recontamination is reversible, provided that the surfaces are exposed to low concentration water at a high flowrate (enhances mass transfer from surfaces to solution). The conclusions from this is that we will use clean-up most of the available time during shutdown to remove activated corrosion products after the addition of hydrogen peroxide.

Table 1 shows a simplified balance of the inventory of gamma source strength in Ringhals 2. It can be seen that roughly 20% of the total gamma source strength from corrosion products are removed in the shutdown transient, and that approximately the same amount remains on the fuel cladding.

It can also be seen that about 50 % of the gamma source strength resides on the stainless steel surfaces. The build-up on the new SG-tubing (1989) is some 10 %.

Table 1. Ringhals 2 shut down 1993. Gamma source strengths on various system parts

System part	Area, m ²	Gamma source strength					
		Mev/s			Fraction of total		
Co-58: 0,97 MeV/Bq		Co-58	Co-60	Sum	Co-58	Co-60	Sum
Co-60: 2,5 MeV/Bq							
SG-TUBING	15315	4,7E+12	5,8E+12	1,1E+13	4%	5%	10%
Stainless steel	2240	1,6E+13	3,6E+13	5,2E+13	15%	33%	48%
Fuel, Zircaloy cladding	4525	1,8E+13	4,9E+12	2,3E+13	17%	4%	21%
Shut down release		2,3E+13	6,8E+11	2,4E+13	21%	1%	22%
Sum		6,2E+13	4,7E+13	1,1E+14	57%	43%	100%
Reducing release/fract of SS		3,4E+12	3,7E+11	3,7E+12	6%	1%	7%
Based on Gammascan of SG, MWI and loop pipes Radiochemical analysis of shut down transient Radiochemical analysis of crud samples from fuel cladding							

Table 2. Ringhals 2 shut down 1987. Gamma source strengths on various system parts

System part	Area, m ²	Gamma source strength					
		Mev/s			Fraction of total		
Co-58: 0,97 MeV/Bq		Co-58	Co-60	Sum	Co-58	Co-60	Sum
Co-60: 2,5 MeV/Bq							
SG-TUBING	14145	2,7E+12	4,8E+13	5,0E+13	2%	41%	43%
Stainless steel	2240	6,4E+12	4,8E+13	5,5E+13	6%	42%	47%
Fuel, Zircaloy cladding	4525	3,0E+12	6,8E+11	3,7E+12	3%	1%	3%
Shut down release		5,9E+12	1,8E+12	7,7E+12	5%	2%	7%
Sum		1,8E+13	9,8E+13	1,2E+14	16%	84%	100%
Based on Gammascan of MWI, pulled tubes 1985 and replaced SG 1989 Radiochemical analysis of shut down transient Radiochemical analysis of crud samples from fuel cladding							

A comparison with 1987 data (before SG replacement) in table 2 indicates that the total gamma source strength is roughly the same as in 1993. However, the amount of Co-60 has been reduced by a factor of almost two. The SG replacement could be considered as a "50 % system decontamination with respect to Co-60" and probably, cobalt as a source for activation.

The 1987 data in table 2 indicates that operation at high pH throughout the cycle minimises the residual activity on the fuel cladding, compared with the modified operation mode employed in 1992 and 1993.

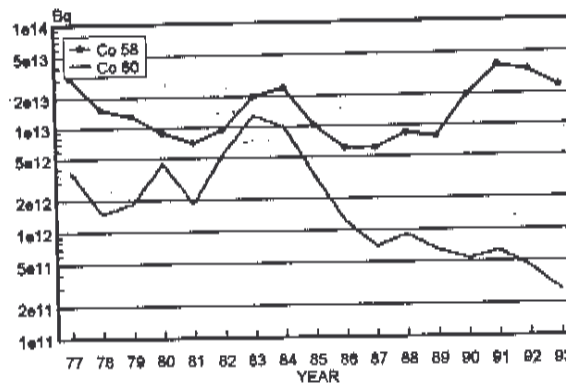


Figure 8. Ringhals 2. Reactor coolant total releases of Co 58 and Co 60 at shut down.

From figure 8 it can be seen that the release of Co-60 at shutdown has decreased over the years. It very probably reflects a decreasing inflow of cobalt to the reactor system. The low releases of Co-58 in 1987-1989 very probably reflects the effect of operation at pH = 7.4, which minimises the solubility and the transport to the core of the corrosion products.

The increasing amounts of Co-58 in the years 1990-1992 probably reflects the corrosion of the fresh SG-tubing material after the SG-replacement in 1989.

FEASIBILITY TEST TO REMOVE FUEL CRUD IN THE FUEL STORAGE POOL.

In the Ringhals PWRs the core is completely unloaded to the spent fuel pool during refuelling. We have observed that the high temperature in the spent fuel pool increases the dissolution rate of activated corrosion products. We have estimated that at 40 °C we may in 3 months remove the same amount of gamma source strength as during the shut down transient.

This method may be used in two different ways: either during prolonged shutdowns, e.g. in connection with SG replacements, or routinely during the cycle to clean the fuel that is to be reloaded into the reactor. The potential of these methods are presently being investigated at Ringhals. Addition of hydrogen peroxide to the fuel pool water will also be tested in combination with the high temperature.

MATERIAL SPECIFICATION TO AVOID HIGH COBALT CONTENT

Ringhals 2 and 3 Replacement Steam Generators

The surface area of the steam generators is very large in relation to the rest of the primary system. We therefore felt that it was important to specify as low Co-content in the steam generator tubing of the replacement steam generators as was technically achievable and economically feasible. In our specification for the new steam generators for Ringhals 2 we required an average Co-content in the Inconel 690 tubing of 0.015 %. This specification was considerably lower than we had in our older generators but also lower than specified in earlier replacement generators. This low Co-content combined with the lower corrosion-rate in Inconel 690 (compared with Inconel 600) and a positive temperature coefficient means a reduction in corrosion products transported to the fuel surfaces.

The tubing for the steam generators for Ringhals 3, which will be replaced in 1995, are specified with a maximum Co-content of 0.02 %. The samples taken indicate an average value below 0.015 %.

Policy for replacement materials

In order to avoid uncontrolled introduction of new stellite components or other high Co-material the following policies are adopted:

- The project department has the responsibility to assure that new Co-sources are not introduced when old systems are modified or new systems installed. Project reports for modifications must include very good motivations if they want to use materials with Co content higher than specified in table 3.
- Stainless steel used in primary systems shall have as low Cobalt content as possible and always below 0.05 %.
- When stellite hard surfaces have to be replaced for functional reasons alternative low Cobalt materials have to be used.
- Strategic materials with low cobalt content shall be stored to avoid the necessity to use what happens to be available on the market.

Fuel Components

Table 3. Cobalt specification for core components.

Components (BWR and PWR)	Material	Maximum Co content
Top end piece	Stainless steel	0.04 % (440 ppm)
Bottom end piece	Stainless steel	0.04 % (440 ppm)
Springs (not spacers) ^a	Inconel	0.04 % (440 ppm)
Centering pins ^a	Stainless steel	0.04 % (440 ppm)
Bolts ^a	Incoloy	0.04 % (440 ppm)
Spacer grids	Inconel	0.02 % (200 ppm)
Spacer grids	Zircaloy	0.001 % (10 ppm)
Fuel cladding	Zircaloy	0.001 % (10 ppm)
Guide thimble tubes (PWR)	Zircaloy	0.001 % (10 ppm)
Fuel channels (BWR)	Zircaloy	0.001 % (10 ppm)

^a Minor deviations can be accepted, provided that total area Co-content as average is less than 0.04 % (440 ppm).

New Low Pressure Turbines in Ringhals 1 (BWR)

A special case is the replacement low pressure turbines ordered for our BWR Ringhals 1. Some of the vendors offering the new turbines wanted to use their standard specification with Stellite surfaces to prevent erosion by the wet steam. Despite full flow cleaning of the condensate the input of cobalt from the turbine system is significant in relation to the contribution from the reactor systems. We therefore managed to include a requirement to have no Stellite blading, except in a very limited number of blades, although the vendor had no experience and therefore was forced to develop an alternative erosion protection.

SUMMARY

As seen in figure 9 collective exposures have been lowered considerably since 1983, when we started our first efforts to optimise our primary chemistry. If you disregard those years with prolonged outages (1989 SG replacement, 1992 and 1993 extensive work on vessel head penetrations) there is a steady reduction in doses. We believe that our high pH and modified chemistry has been of importance but of course we have also spent a lot of efforts to develop working methods, robotics, training etc. We have also been able to reduce the normal outage from 50 to 75 days ten years ago to 27 to 35 days today and avoid forced outages by improved maintenance programs. All these factors do of course contribute to the results we have achieved.

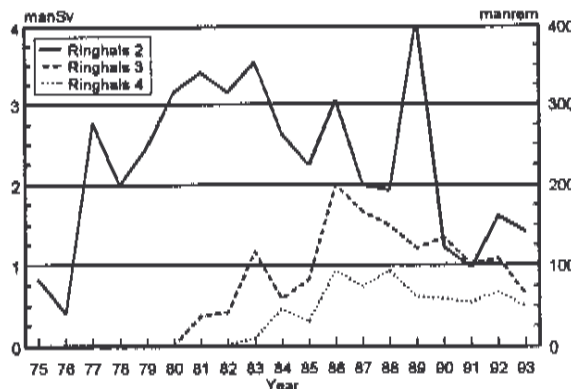


Figure 9. Ringhals 2, 3 and 4 annual collective exposures.

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Author Biography

Krister Egnér is Manager of the Environment and Safety Department at the Ringhals Nuclear Power Plant. For 15 years he has been the licensed Radiological Supervisor for the Ringhals site and a member of the Ringhals Executive Group. He has been the Swedish representative in the discussions leading to the formation of the ISOE system and, until recently, a member of the ISOE Steering Committee. He is the Chairman of the Reactor Safety Review Committee for the Ringhals site and a member of the Vattenfall Corporate Reactor Safety Review Committee. He has a M.Sc. in Mechanical Engineering from Chalmers University of Technology in Gothenburg, Sweden.

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