

CONTROLLING RADIATION FIELDS IN SIEMENS DESIGNED LIGHT WATER REACTORS

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ABSTRACT

An essential item for the control of radiation fields is the minimization of the use of stellites in the reactor systems of Light Water Reactors (LWRs). A short description of the qualification of Co-replacement materials will be followed by an illustration of the locations where these materials were implemented in Siemens designed LWRs. Especially experiences in PWRs show the immense influence of reduction of cobalt sources on dose rate buildup. The corrosion and the fatigue and wear behavior of the replacement materials has not created concern up to now.

A second tool to keep occupational radiation doses at a low level in PWRs is the use of the modified B/Li-chemistry. This is practiced in Siemens designed plants by keeping the Li level at a max. value of 2 ppm until it reaches a pH (at 300°C) of ~7.4. This pH is kept constant until the end of the cycle.

The substitution of cobalt base alloys and thus the removal of the Co-59 sources from the system had the largest impact on the radiation levels. Nonetheless, the effectiveness of the coolant chemistry should not be neglected either.

Several years of successful operation of PWRs with the replacement materials resulted in an occupational radiation exposure which is below 0.5 man-Sievert/plant and year.

INTRODUCTION

Radiation Field Control in Siemens designed Light Water Reactors (LWRs) will be the continuation of the efforts in the last 20 years. An example for these efforts: the average annual radiation exposure in Siemens designed PWRs was 5.25 man-Sievert per plant and year in 1980 and 1.73 man-Sievert per plant and year in 1990. A comparison of these values with statistics from other countries shows that the average personnel exposure of all Siemens plants is comparable to the values in most other countries.

However, taking into account only Siemens plants, which started operation after 1985, a decrease of personnel exposures of one order of magnitude can be observed compared to those plants which started operation before 1985. In order to achieve personnel exposures in such a low range, new concepts in shielding and material selection of these "recent" plants were necessary. The most important objective hereby was to reduce the radiation levels. To obtain this objective, the Co-60 had to be eliminated by eliminating its precursor Co-59.

Therefore a new material concept had to be developed. In order to enable the realization of this concept, the main cobalt sources had to be identified and suitable replacement materials had to be

qualified. By using the new qualified materials a stepwise reduction of the identified relevant cobalt sources could be performed.

Simultaneously, a new concept for primary coolant chemistry was developed. The intention hereby was to achieve a pH in the primary coolant which would have a stabilising effect on the oxide layers. Especially the re-dissolution of nickel and cobalt and their transport was to be minimized.

IMPROVEMENT OF THE MATERIAL CONCEPT

During extensive research programs in the 70s, cobalt base alloys with a cobalt content of > 50%, especially Stellite 6, used in the reactor pressure vessel (RPV) area, in valves, and in pumps were identified as main cobalt sources. Furthermore, plating material and fuel assembly materials, located in the neutron field of the RPV and containing cobalt as an impurity, were taken into consideration as a Co-60 source.

Thus, the development and testing of replacement materials was started in the 70s, considering mainly the core internals made of cobalt base alloys, and was continued in the 80s, taking into account then the entire primary system. A list of replacement materials with a significantly reduced cobalt content (Table 1) and various mechanical characteristics (Figure 1) resulted. The replacement of most cobalt base alloys was now enabled by selecting these materials according to the location and mechanical load of the relevant components.

Table 1. Chemical composition of Co-free replacement alloys

ALLOY	Hardness HRC	Chemical composition in weight %									
		C	Si	Mn	Cr	Mo	Ni	N	W	V	Fe
EVERIT 50	47-54	2.5	0.4	0.9	25	3	-	-	-	-	bal.
EVERIT 50 SO	43-48	2.0	0.4	0.9	25	3	-	-	-	-	bal.
ANTINIT DUR 300	27-33	0.12	5	6.5	21	-	8	-	-	-	bal.
CENIUM Z 20	43-48	0.3	-	-	27	9	17.5	-	2	-	bal.
SKWAM	36-42	0.2	0.7	0.55	17.5	1.1	-	-	-	-	bal.
NITRONIC 60	20-29	0.1	4	8	17	-	8	0.13	-	-	bal.
Cr-STEELS	24-44	0.2	<1	<1	16.5	-	2	-	-	-	bal.

Detonation-gun-coatings: Coating type LC-1C of union carbide
Composition Cr₃C₂ with 20% Ni-Cr binder

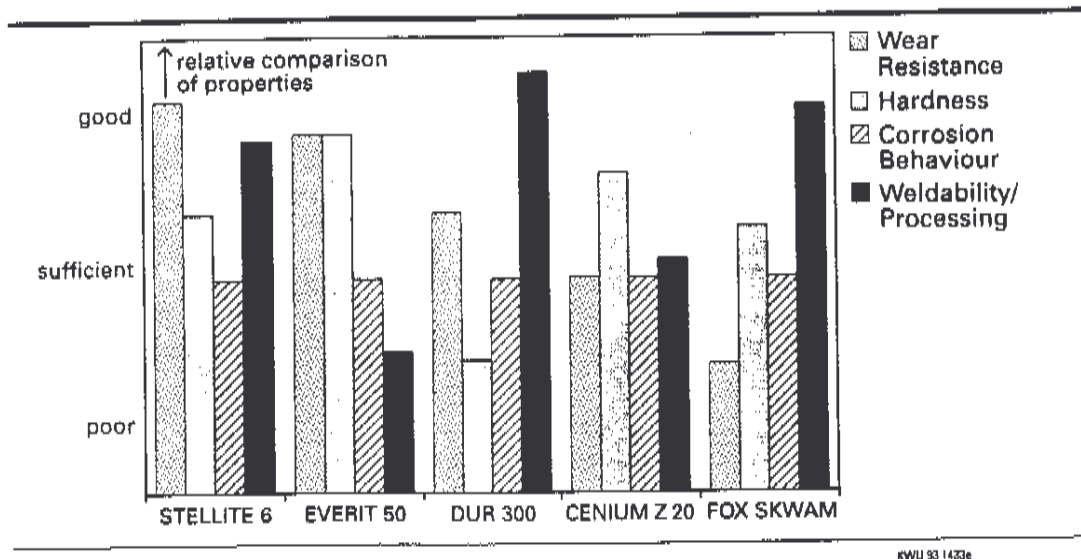


Figure 1. Comparison of main properties of hardfacing alloys

In parallel, the guide tubes, manufactured from stainless steel, and the spacer grids, manufactured from Inconel, were replaced during refuelling outages by those made from Zircaloy.

The first pressurized water reactor (PWR) designed by Siemens with Co-replacement materials started operation in 1985. At this PWR only stellites located in-core were replaced by "cobalt-free" materials. In the following plant the stellites located at the main coolant pumps and the main valves were included in the replacement program. All valves, even those in the auxiliary systems, were taken into account only in the Konvoi plants.

The transition from "older" to "recent" 1300 MWe Siemens designed PWRs can be quantified by comparing the surface areas of the individual materials listed in Tables 2 and 3. As additional information, these tables show the specified Co-59 content of the materials.

Table 2. Materials inventory of "older" 1300 MWe Siemens-designed PWRs

Group	Component	Material	Surface [m ²]	Co-59 Specification [%]
1	Fuel*)	Zircaloy 4	9600	-0
		Inconel 718	1220	<0.1
		Stainless steel	220	<0.1
	RPV-internals	Stainless steel	1124	<0.1
		Co-base alloys	1.1	63
2	Control rod assemblies	Stainless steel	340	<0.1
	Control rod drive	Stainless steel	220	<0.2
		Co-base alloys	1.54	≤67
	Steam generator	Incoloy 800	16276	<0.1
	RPV, Loops	Stainless steel	719	<0.2
	Main coolant pumps	Stainless steel	155	<0.2
Co-base alloys		1.5	63	
3	Auxiliary systems	Stainless steel	~500	<0.2
		Co-base alloys	6.5	63
	Total	Zircaloy	9660	
		Stainless steel	19554	
		Inconel	1220	
		Co-base alloys	10.64	

*) Material composition used before 1985, modifications per fuel cycle possible

Table 3. Materials inventory of "recent" 1300 MWe Siemens-designed PWRs

Group	Component	Material	Surface [m ²]	Co-59 Specification (%)
1	Fuel*)	Zircaloy 4	-10660	<0
		Inconel 718	394	<0.1
		Stainless steel	220	<0.1
	RPV-internals	Stainless steel	1126	<0.1
	Co-base alloys	0.026	63	
2	Control rod assemblies	Stainless steel	340	<0.1
	Control rod drive	Stainless steel	220	<0.2
		Co-base alloys	1.54	≤67
	Steam generator	Incoloy 800	16276	<0.1
	RPV. Loops	Stainless steel	719	<0.2
	Main coolant pumps	Stainless steel	156	<0.2
Co-base alloys		0	63	
3	Auxiliary systems	Stainless steel	508	<0.2
		Co-base alloys	0.79	63
	Total	Zircaloy	10660	
		Stainless steel	19563	
		Inconel	394	
		Co-base alloys	2.36	

*) Modifications per fuel cycle possible

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The components in these tables are divided into three groups depending on their location:

- Group 1: Contains only components which are permanently in the neutron flux
- Group 2: Contains components which are outside the neutron flux but within the main circuit
- Group 3: Contains components from the auxiliary systems

Approx. 98 % of the cobalt base alloys located in-vessel were substituted by "cobalt-free" materials as can be seen when comparing the surface areas of Group 1. A reduction from 1.1 m² in "older" plants to 0.026 m² in "recent" plants was achieved. This in-vessel replacement had the greatest impact on radiation levels according to the fact that some of these hardfacings were placed close to the neutron flux area.

A comparison of the material concepts in Group 2 shows that the cobalt base alloys of the control rod drives were not replaced. This decision was based on the fact that the cobalt hardfacings in this case have almost no contact to the primary coolant. The release rates of Co-59 from this component into the coolant therefore cannot be very high. Nevertheless, the cobalt base alloys of the main coolant pumps were removed entirely. Thus, a reduction of approx. 50 % of the surface areas was achieved with an effectiveness much higher than 50 %, considering the Co-59 release into the coolant and hence the potential Co-60 buildup rate.

A major reduction (approx. 88 %) of the surface areas from hardfacings was also obtained in the auxiliary systems. However, the influence on the radiation levels was much less here than in Group 1 and also less than in Group 2, because of the reduced contact between these auxiliary systems and the primary circuit.

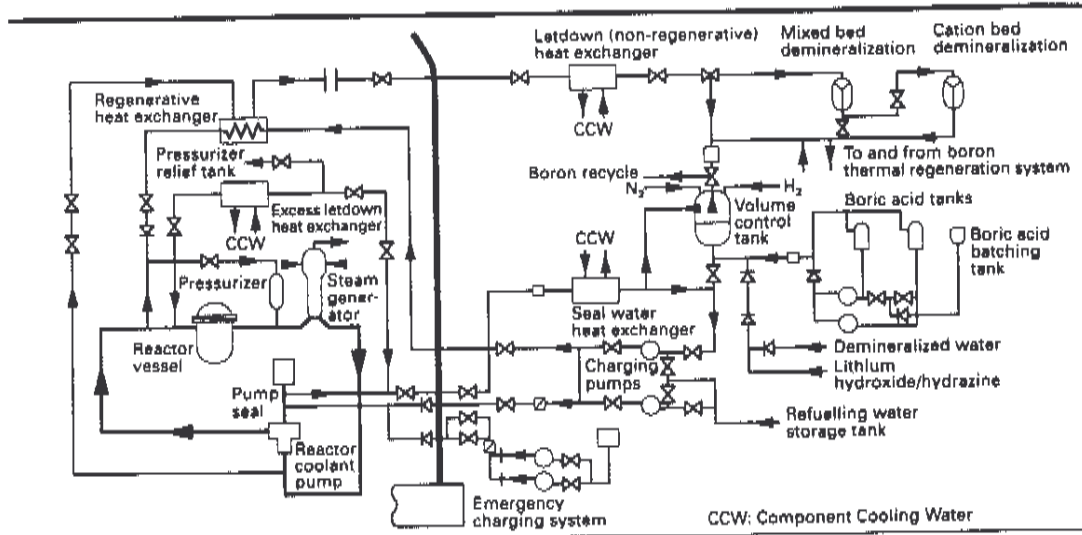


Figure 2. Example of main coolant circuit with CVCS
(IAEA, Technical Reports Series No. 347, Vienna, 1993)

The schematic diagram in Figure 2 shows the chemical and volume control system (CVCS) and its connections to the primary circuit.

The replaced in-vessel components (Group 1) are shown in more detail in Table 4 and Figures 3 and 4, since the highest effectiveness on the reduction of the radiation levels was gained by these replacements. The table is valid for substitutions in Philippsburg-2 as well as in the Konvoi plants. The figures have been derived from Philippsburg-2.

Table 4. Substituted in-vessel stellites in Philippsburg-2 and Siemens Konvoi plants

Component	Amount	Gross Area, [m ²]	Substitute
Alignment pin with nut for support columns (control rods)	244	0.8	Cr ₃ C ₂ / CrNi-binder
Grid plate centering bolts	4	0.08	Fox Antinit Dur 300
Hold down plates of the upper core support	112	0.24	Fox Antinit Dur 300
Centering bolts of the upper core support	4	0.04	Fox Antinit Dur 300
Total substituted		1.16	
Total Remaining		0.026	

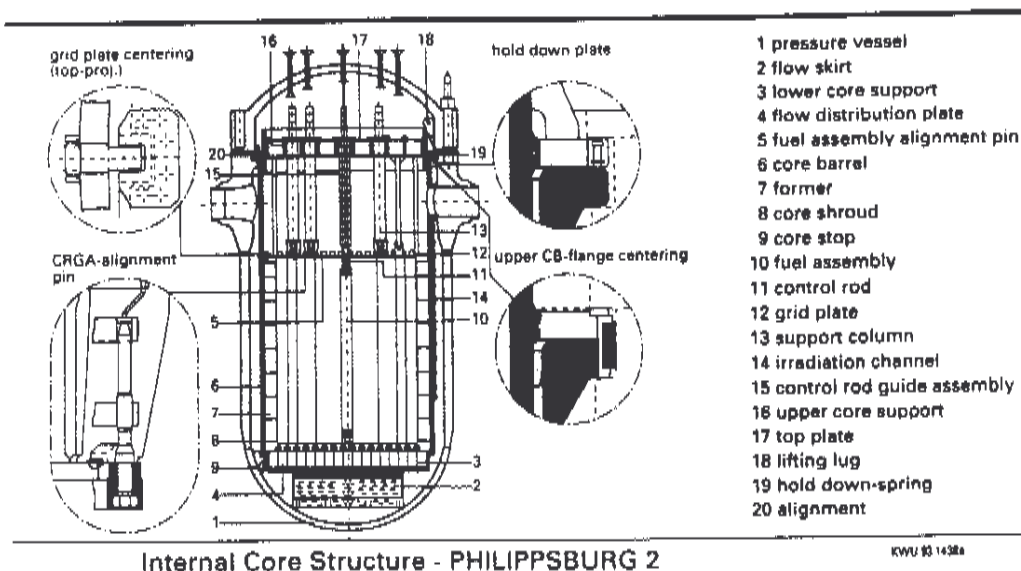


Figure 3. Reactor pressure vessel internals and fuel assemblies

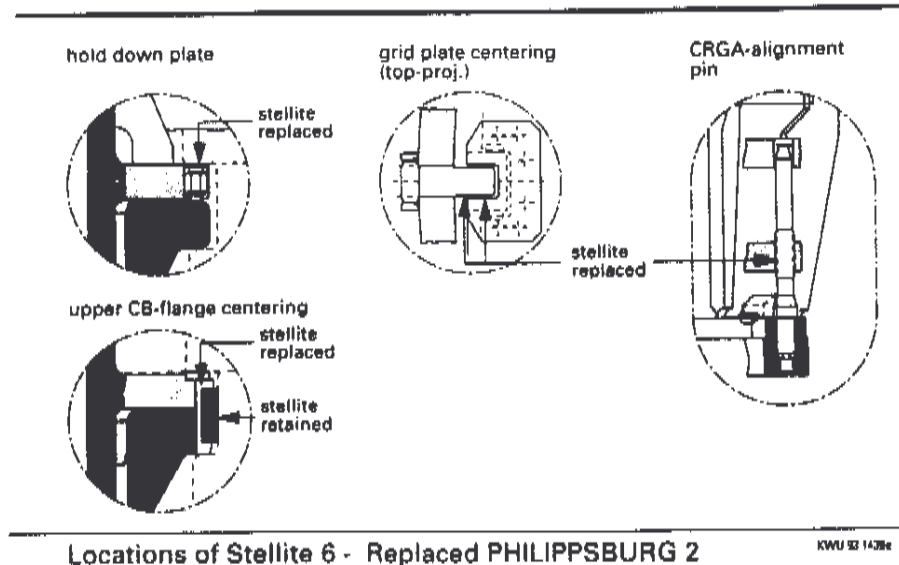


Figure 4. Reactor pressure vessel internals and fuel assemblies

MODIFICATION OF PRIMARY COOLANT CHEMISTRY

The concept of the coordinated coolant chemistry, pH (300 °C) = 6.9, was based on the assumption that the oxide layers in the primary circuit mainly consist of magnetite. However, further research work has shown that oxide layers as well as crud in the primary circuit are mainly composed of spinels in which the Fe(II) of the magnetite is substituted by other bivalent cations. Nickel and cobalt, respectively their activation products Co-58 and Co-60, are thus incorporated in the oxide layers and thereby increase the radiation field.

Considering this and knowing furthermore that, depending on the composition of the Ni- and Co-ferrites, the minimum solubility of these ferrites lies at a pH-level in the range of 7.4, a primary coolant chemistry different from the previous one must be recommended.

Preferably a chemistry with a constant pH, selected from the above mentioned range, should be chosen. However, the lithium concentration is limited to 2 ppm. Otherwise the material compatibility especially of fuel element material is no longer guaranteed. Since lithium hydroxide is used as the pH control agent, a compromise must be found. Therefore it is recommendable to first adjust a constant lithium concentration in the primary coolant until the desired pH is achieved and then to reduce the lithium concentration in dependence upon the boron concentration in such a way that a constant pH can be applied until the end of the cycle.

The effectiveness of this modified coolant chemistry can be experienced by comparing the dose rate development of plant A and plant B in Figure 5. Stellite replacement was not performed in either of these two plants, so that they differ mainly in the primary coolant chemistry. In plant A, with the higher dose rate, coordinated coolant chemistry was preferred with $\text{pH}(300) = 6.9$, whereas in plant B modified coolant chemistry was applied from the very beginning, in this case $\text{pH}(300) = 7.4$.

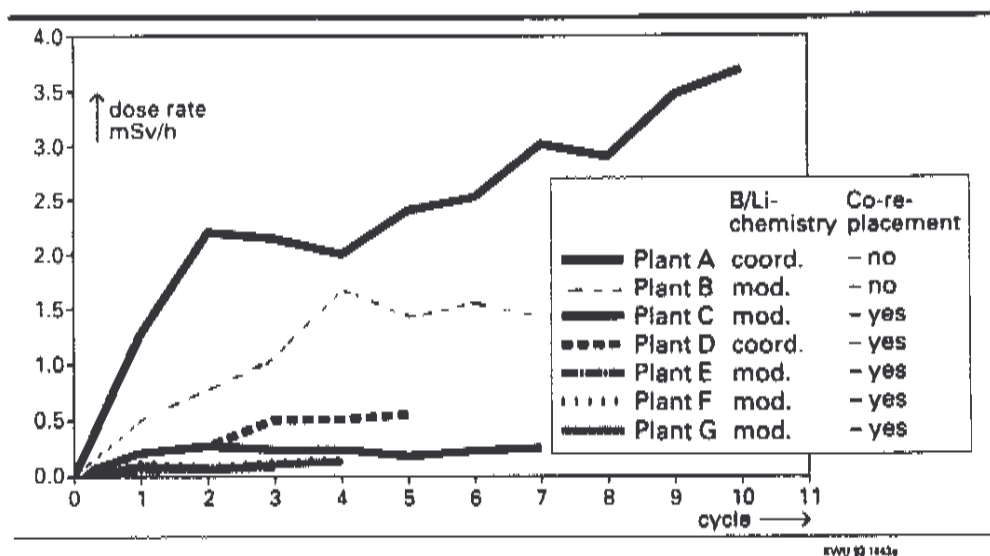


Figure 5. Dose rate development at main coolant piping of seven Siemens PWRs

RADIATION FIELD DEVELOPMENT

As a consequence of these measures taken, the radiation fields and the average occupational radiation exposures decreased in the Siemens designed PWRs. Figure 6 shows this development by classifying the PWRs into 3 groups.

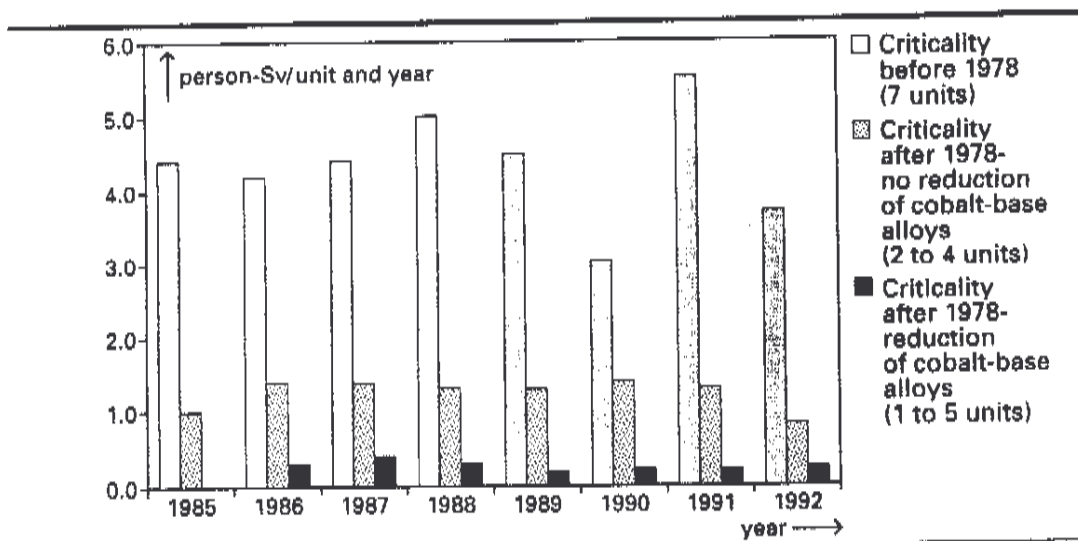


Figure 6. Average occupational radiation exposure per unit and year of Siemens PWRs

The PWRs in the first and second group had no reduction of cobalt base alloys. However, the plants in the first group (oldest plants), which had their first start-up before 1978, had generally uncoordinated or coordinated coolant chemistry during the early fuel cycles and only poor shielding, whereas the plants in the second group, which started first operation after 1978, applied mainly modified coolant chemistry from the very beginning and also had improved shielding. Yet, the third group consists of plants at which the cobalt base alloys were substituted either partly or in full extent as described in Table 3. This last group shows that the measures performed succeeded in an average occupational radiation exposure of less than 50 Rem per plant and year.

Figure 7 is an example for the dose rate development of various PWRs with Co-replacement activities but different coolant chemistry.

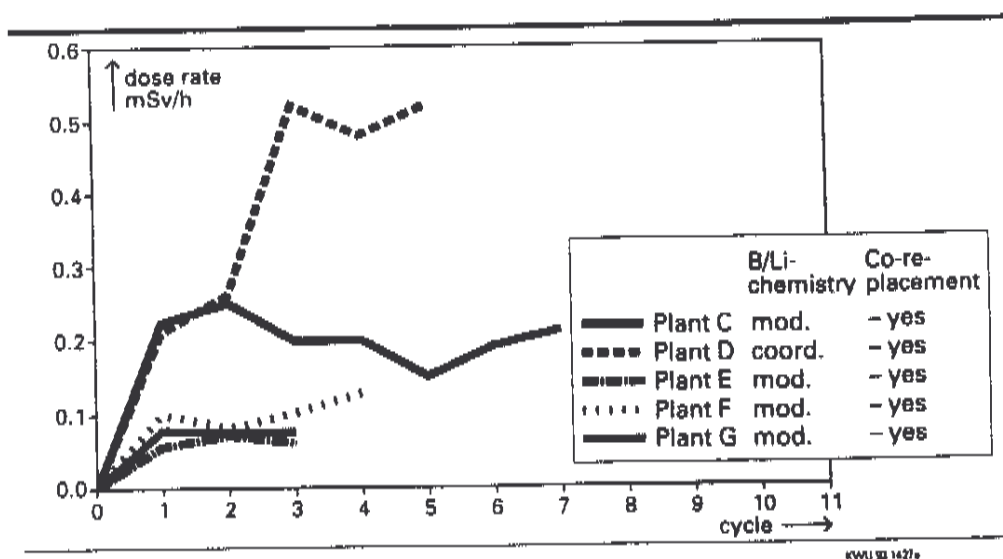


Figure 7. Dose rate development at main coolant piping of five Siemens designed PWRs

Compared to Figure 5, this figure shows that the influence of the coolant chemistry on the radiation levels is not as large as that of material replacement, but it still should not be neglected.

The overall objective of Co-replacement activities can be illustrated with Figure 8, where a comparison of soluble elemental cobalt-59 in the primary coolant is shown in comparison with channel head dose-rates. According to this figure the overall objective should be to reduce the soluble Co-59 to values less than 5 ppb in order to thus achieve distinctly reduced dose rate levels.

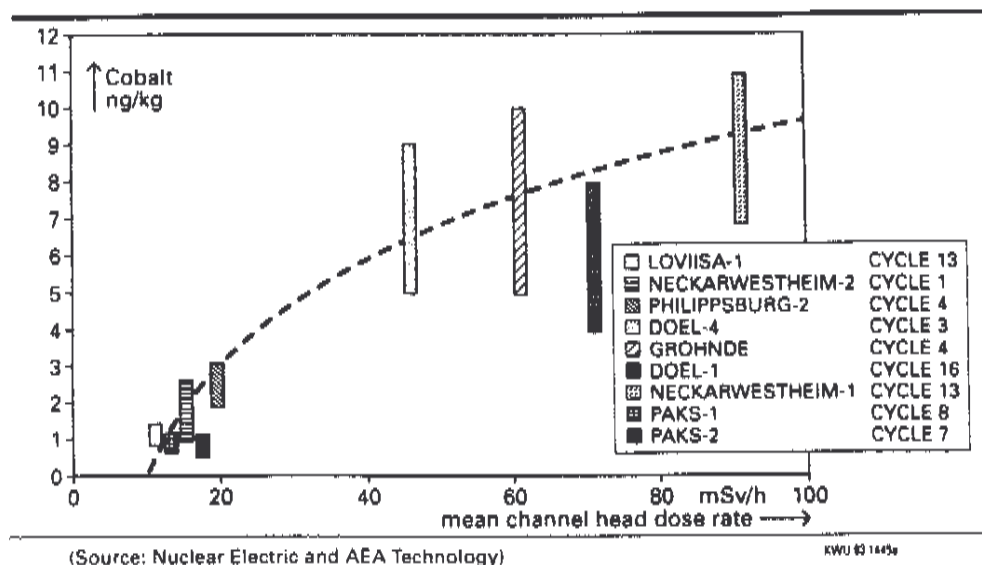


Figure 8. Comparison of soluble elemental cobalt in the primary coolant with channel head dose rate

Author Biography

Rolf Riess is Senior Director for the Power Plant Chemistry Division of Siemens AG KWU in Erlangen, Federal Republic of Germany. The division is responsible for all aspects of Power Plant chemistry including research and development, and service activities like radiation control, decontamination and steam generator chemical cleaning. Before joining Siemens, Dr. Riess was a scientist for one year at the Institute for Nuclear Chemistry at the Technical University in Darmstadt, FRG. He received a Ph.D. degree in Chemical Engineering from the Technical University of Darmstadt, Federal Republic of Germany.

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PAPER 3-3 DISCUSSION

Khan: Rolf, you've shown that the doses for your plants are now extremely low. Do you think that we have reached a kind of a bottom line, or is there any more possibility of reducing doses even further in Siemens plants? Question number two, what kind of pH are you operating at in your newer plants?

Riess: As I said, our intent is to further reduce these exposure rates by those measures I mentioned. Take counteractions against nickel, that is the main target, further reduce the cobalt in the auxiliary systems, mainly in the SVCS equipment, of of course, provide decontamination as a routine measure against residual fields if you have to do specific works. These are the major actions that we have on the way. As to the second question about the pH, we introduced another feature about 10 years ago. We are operating at the upper level of the lithium concentration, in our case it is 2 ppm of lithium up to 7.4, and then follow this line. So except for three stations out of the 16, all are following the modified chemistry. Admittedly, this is a bit of a gamble, but we think we are in pretty good shape.