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THE IMPACT OF STEAM GENERATOR REPLACEMENT ON PWR PRIMARY SYSTEM CONTAMINATION

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ABSTRACT

This paper analyses the impact of Steam Generator Replacement (SGR) on PWR primary circuit contamination. It presents a comparison of the activities deposited inside the primary system and released during refuelling outages after SGR with three different SG tube alloys (600, 690 and 800) and different SG tube manufacturing processes.

A SGR has a great impact on the primary system contamination. After SGR, whatever the SG tube material is, the typical variations are the following:

- The ^{58}Co contamination increases for 1 to 3 cycles, and then decreases to very low levels in some cases, mainly depending on the manufacturing process of the replacement SG tubes.
- The ^{60}Co contamination tends to decrease on the primary coolant pipes and increases by a lower rate on the new SG tubes.

This analysis highlights the importance on contamination levels after SGR of both the corrosion product deposits on the primary surfaces before SGR and the surface finish of the SG tubes related to their manufacturing process.

1. INTRODUCTION

In 2000, the previous paper dealing with the impact of Steam Generator Replacement (SGR) on PWR primary system contamination explained that the two main key factors were the corrosion product deposits before SGR and the manufacturing process of the replacement SG tubes [1]. Ten years later, the SGR influence on PWR contamination has been reassessed with enriched experimental feedback.

Two radioactive isotopes, ^{58}Co and ^{60}Co , contribute by over 90% on the average to the dose rates around primary systems [2]. They come from the activation of nickel and cobalt, according to: ^{58}Ni (n,p) ^{58}Co (radioactive half-life: 71 days) and ^{59}Co (n, γ) ^{60}Co (radioactive half-life: 5.3 years).

As shown in Figure 1, ^{58}Co is predominant in dose rates for the first reactor cycles. This figure presents the $^{58}\text{Co}/^{60}\text{Co}$ ratio of primary system surface activity measured by gamma spectrometry using EMECC devices [3] in about 60 different PWRs. The ^{58}Co and ^{60}Co contributions to the dose rates are equivalent in the green band. Progressively, the ^{58}Co contribution decreases giving way to ^{60}Co , which accumulates due to its longer radioactive half-life. Nevertheless for some PWRs, after several cycles, (red triangles in Figure 1), the ^{58}Co contribution reaches the level of the first cycles again. This increase is the result of SGR.

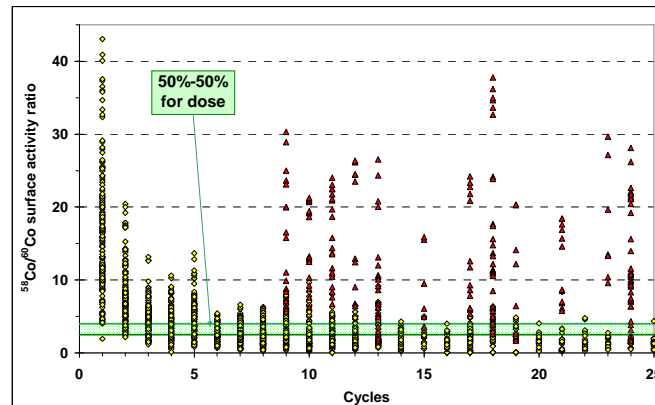


Figure 1. $^{58}\text{Co}/^{60}\text{Co}$ surface activity ratio in PWR primary systems.

Because of Inconel 600 Mill-Annealed tube corrosion problems (intergranular attack, stress corrosion cracking ...), PWR steam generators must be replaced. The material and the manufacturing process of the replacement SG tubes vary: namely, Thermally-Treated Inconel 600, Inconel 690TT or Incoloy 800.

Regarding contamination, it is expected that:

- The heat treatment of Inconel should reduce the release [4],
- The higher Chromium content of Inconel 690 (about 29% Cr, 59% Ni, 10% Fe) should reduce the generalized corrosion rate,
- The lower Nickel content of Incoloy 800 (about 33% Ni, 21% Cr, 44% Fe) should reduce the ^{58}Co source.

The impact of these tube materials on contamination is compared and the typical evolutions of ^{58}Co and ^{60}Co contamination due to SGR are described and analyzed in the following sections.

2. DESCRIPTION OF UNITS

In order to study the effect of SGR in the contamination of PWR primary systems, CEA, within the framework of EDF and other operators projects, performed in-situ gamma spectrometry measurements using the EMECC device [3] in 18 PWRs, before and after SGR, both in France and in other countries. Three types of replacement SGs were studied:

- 15 SGRs with alloy 690TT tubes manufactured with different processes,
- 1 SGR with alloy 600TT tubes,

- 2 SGRs with alloy 800 tubes.

The EMECC measurements lead to an accurate characterization of the contamination levels (surface activity by activated corrosion product) at the time of some refueling outages.

In this paper, we focus on the 5 most representative SGRs. For each of them, Table 1 shows:

- The unit considered.
- The SGR outage (cycle number).
- The year of the SGR outage.
- The alloy of the replacement steam generator tubes. The alloy of the replaced SG tubes in all units was Inconel 600MA.
- The steam generator wet surface area before and after SGR.

Table 1. Description of the SGRs.

Unit	SGR outage	SGR year	RSG tube alloy	SG wet area Bef./Aft. SGR (m ²)
A	12	1993	600TT	4400/4400
B	8	1990	690TT	4400/4400
C1	11	1995	690TT	4400/4400
C2	19	2003	690TT	4400/5000
D	11	1993	800	4200/5700

These units are 900 MWe PWRs. For unit D, the power was increased by about 15% after SGR.

The first operating cycle with a coordinated chemistry program at $\text{pH}_{300^{\circ}\text{C}} = 7.2$ and a maximum lithium concentration of 2.2 ppm was cycle 17 for unit A, cycle 16 for unit B and cycle 14 for units C1 and C2. For the previous cycles, the target $\text{pH}_{300^{\circ}\text{C}}$ was 7.0 with 2.2 ppm maximum lithium.

The manufacturing processes of the Inconel 690TT SG tubes of units B, C1 and C2 differ from the oldest to the most recent respectively.

There was no particular preconditioning at the end of the SGR outage of these units in order to “passivate” the replacement SG tube surfaces

3. VARIATIONS IN CONTAMINATION

3.1 ⁵⁸Co surface activity

The ⁵⁸Co surface activity inside the primary system (hot legs, crossover legs and steam generator tubes at about 2 m above the tubesheet) of units A, B, C1, C2 and D is presented in Figure 2 (primary coolant pipes) and Figure 3 (SG tubes – C: Cold side / H: Hot side).

Whatever the material of the replacement SG tubes (600, 690 or 800) and their manufacturing process are, the ⁵⁸Co surface activity generally increases for 1 or 2 cycles inside the primary coolant pipes after SGR (see Figure 2). Afterwards they decrease and, after several cycles, they reach similar levels or even lower levels than those prior to SGR.

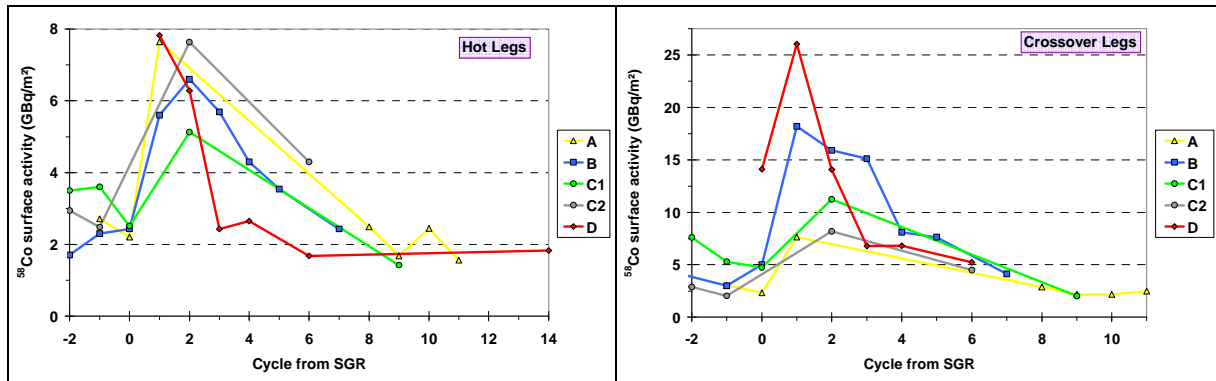


Figure 2. ⁵⁸Co surface activity inside the primary coolant pipes.

For the replacement SG tubes, two types of behavior have been observed (see Figure 3):

- The ⁵⁸Co surface activity increases for 1 to 3 cycles after SGR and then decreases and reaches similar or lower levels than those recorded prior to SGR (units B, C1, C2 hot side and D);
- The ⁵⁸Co surface activity seems to increase throughout more than 3 cycles and then stabilizes at levels as high as those prior to SGR (units A and C2 cold side).

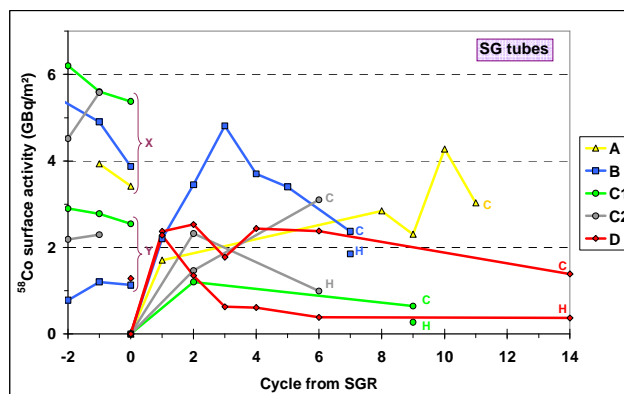


Figure 3. ⁵⁸Co surface activity inside the SG tubes.

The high level of ⁵⁸Co contamination on the SG cold side of unit C2 was unexpected. The ⁵⁸Co surface activity is about 3 times higher than that in unit C1 and similar to unit B and even unit A (see Figure 3). Yet, units C1 and C2 are twin units of a same nuclear power plant and they operate under the same conditions: cycles of 10 to 12 months, coordinated chemistry at pH_{300°C} = 7.0 until cycle 13 and afterwards at pH_{300°C} = 7.2. Furthermore, all the fuel assemblies were equipped with Zircaloy grids before SGR and the Inconel 600MA SG tubes were replaced by Inconel 690TT SG tubes. On the other hand, the differences between the two SGRs are the following:

- The SGR outage, cycle 11 for unit C1 and cycle 19 for unit C2. Nevertheless, the ^{58}Co (and ^{60}Co) contamination had reached equilibrium before SGR in unit C2 and in unit C1 as well.
- The SG surface area increased by 14% in unit C2 after SGR whereas the Inconel 690 surface area is the same as Inconel 600 in unit C1. However, the release SG surface area is higher but the deposition SG surface area is as well.
- The only identified difference which could explain the high ^{58}Co level of the cold side SG tubes is the manufacturing process of the Inconel 690TT tubes that has changed between unit C1 and unit C2. Apparently, the SG tube manufacturing process in unit C2 led to a higher corrosion product release than that in unit C1.

The major effect of the SG tube manufacturing process may also be observed before SGR. In Figure 3, we have distinguished the EMECC measurements of SG tubes before SGR as a function of the 600 alloy tube manufacturer “X” or “Y” (e.g. in unit B, the 600 SG tubes of loops 1 & 3 were manufactured by “X” and those of loop 2 by “Y”). For each unit B, C1 and C2, all the SGs obviously were subjected to the same conditions (water chemistry, temperature, fluid velocity...) and yet the SG tubes manufactured by “X” were about 2 to 4 times more contaminated than those manufactured by “Y”. The SG tubes manufactured by “Y” released less corrosion products and trapped less contamination [2].

A characteristic of the replacement SG tubes is the fact that the differences in contamination between the hot side (H) and the cold side (C) of the SG tubes are marked in units C1, C2 and D (see Figure 3). The hot side is less contaminated than the cold side, by a factor of 2 to 6. This may be due to a cobalt equilibrium concentration lower at 280°C (cold side) than at 320°C (hot side) in relation with the SG tube oxides. However, the case was the opposite 2 cycles after SGR in unit C2, i.e. the hot side was more contaminated than the cold side (by about +60%). The SG tube oxide structure likely changed during cycles after SGR. This inversion was also observed in other units after SGR. Note that before SGR, there was practically no difference in contamination between the two sides of the SG tube bundles.

Although the replacement SG tube alloy of unit D is Incoloy 800, i.e. with a lower Ni content (about 33% vs. about 59% for Inconel 690), the ^{58}Co surface activity inside the primary system of unit D is similar or even higher than that observed after SGR with Inconel 690TT, a situation that was unexpected. 14 cycles after SGR, the Incoloy 800 release was still high.

It should be noted that the replacement of Inconel 718 spacer grids with Zircaloy spacer grids from SGR outage in unit A contributed to a reduction of the contamination [5], as well as for the first refueling outage after SGR in unit B

3.2 ^{60}Co surface activity

The ^{60}Co surface activity inside the primary system (hot legs, crossover legs and steam generator tubes) of units A, B, C1, C2 and D is presented in Figure 4 (primary coolant pipes) and Figure 5 (SG tubes – C : Cold side / H : Hot side).

After SGR, no matter what the type of replacement SG tube alloy used, the ^{60}Co surface activity tends to decrease or at least remain constant inside the primary coolant pipes (see Figure 4). Such is also the case for the ^{58}Co contamination (see §3.1), the ^{60}Co contamination decrease after SGR in units A and B is partly due to the replacement of the Inconel 718 spacer grids by Zircaloy spacer grids.

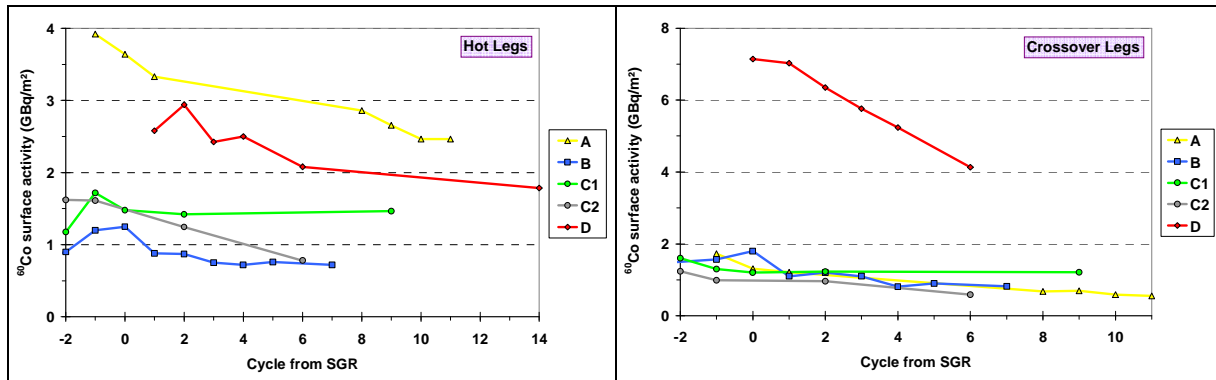


Figure 4. ⁶⁰Co surface activity inside the primary coolant pipes.

Inside the replacement SG tubes, the ⁶⁰Co surface activity increases for several cycles and generally stabilizes at a lower level than that seen prior to SGR, except for the SG cold side tubes of unit D (see Figure 5). Unit D is characterized by a high level of ⁶⁰Co contamination, which was already the case before SGR and which was due to an abnormal wear of Stellites or the use of Inconel 718 spacer grids having a large Co content [2]. Such is also the case for ⁵⁸Co (see §3.1), the SG hot side is less contaminated by ⁶⁰Co than the cold side, particularly in unit D.

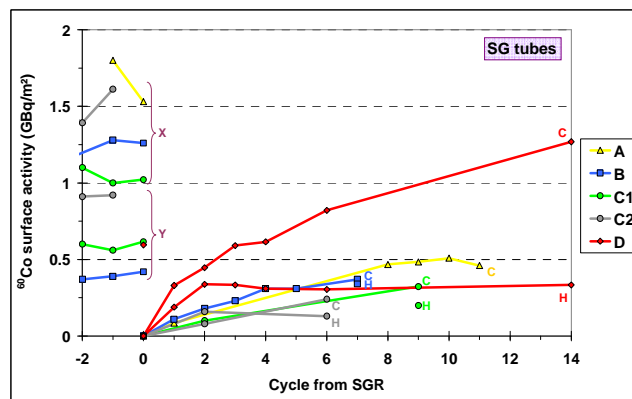


Figure 5. ⁶⁰Co surface activity inside the SG tubes.

3.3 Dose rates

The dose rate index of units A, B, C1 and C2 is presented in Figure 6. The dose rate index is an average of dose rates measured at the primary coolant pipes by the operator (hot leg, crossover leg, cold leg of each loop).

Except for unit A, the dose rate index generally increases for 1 or 2 cycles after SGR and then decreases. The high value at SGR outage of unit A shows that a dose rate measured in contact with a primary coolant pipe is not only representative of the actual surface activity of the primary

coolant pipe but it is also and especially representative of the ambient dose rate (e.g. hot spot inside an auxiliary pipe, volume activity...).

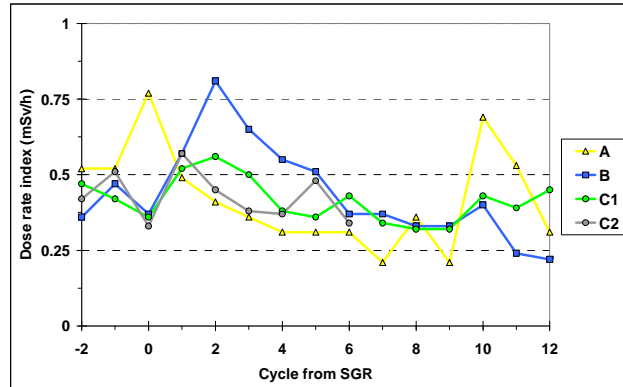


Figure 6. Dose rate index.

The dose rate variation is primarily due to the variation in the ^{58}Co surface activity inside the primary coolant pipes (see Figure 2). Thus, the ^{58}Co contribution to the dose rates around the primary coolant pipes increases after SGR and can be the major one for several cycles to the detriment of the ^{60}Co contribution as shown in Figure 7. Figure 7 presents the ^{58}Co and ^{60}Co average contributions to the dose rates around the primary coolant pipes. Thanks to the MERCURE code [6], the radionuclide contributions were calculated on contact with the thermal insulations of the primary pipes by using surface activity and component characteristics.

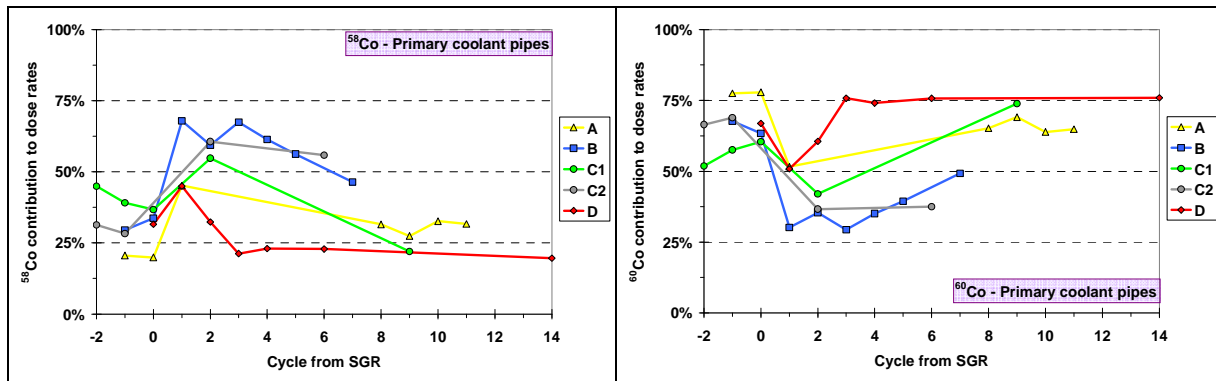


Figure 7. ^{58}Co and ^{60}Co contributions to the dose rates around the primary coolant pipes.

Similarly, ^{58}Co is predominant in dose rates around the SG shells for the first cycles after SGR (see Figure 8). Progressively, the ^{58}Co contribution decreases giving way to ^{60}Co , which accumulates due to its radioactive half-life.

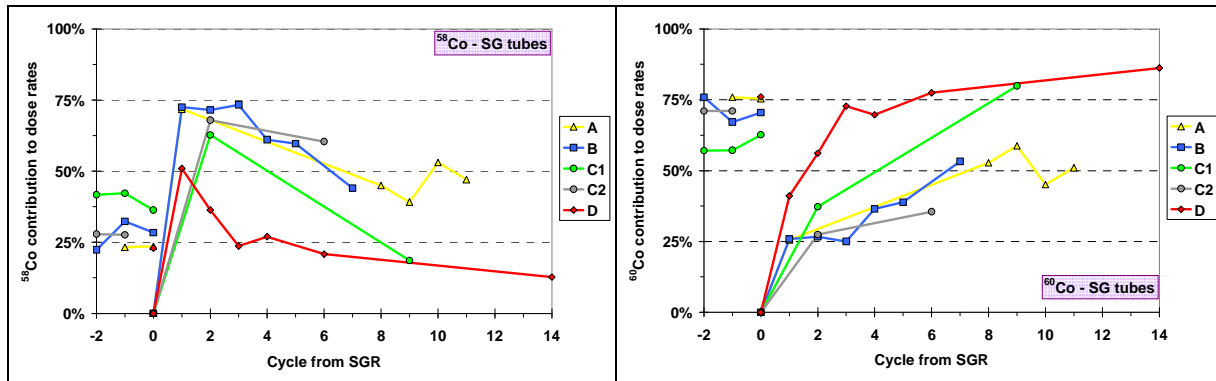


Figure 8. ⁵⁸Co and ⁶⁰Co contributions to the dose rates around the SG shells.

3.4 Peak ⁵⁸Co activity during refueling outages

Figure 9 presents the peak ⁵⁸Co activity after the primary coolant oxygenation during the refueling outages in units A, B, C1, C2 and D.

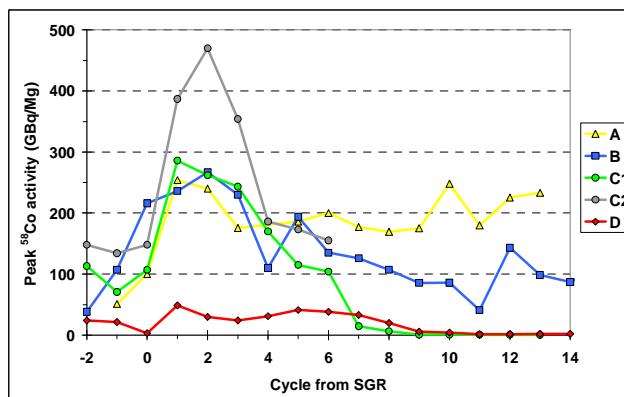


Figure 9. Peak ⁵⁸Co activity after the primary coolant oxygenation during refueling outages.

As for the ⁵⁸Co surface activity (see §3.1), the peak ⁵⁸Co activity after the primary coolant oxygenation increases for 1 or 2 cycles after SGR and then decreases. Before SGR, the peak ⁵⁸Co level due to the primary oxygenation is about 100 GBq/Mg. The first two or three cycles after SGR, it is usually higher than 200 GBq/Mg and it can even reach about 500 GBq/Mg. Note that the shutdown procedure had not changed after SGR for units A, B, C1 and C2 [7], and the primary coolant oxygenation occurred before the draining of the reactor coolant system.

Several cycles after SGR in unit A (Inconel 600TT RSG), the peak ⁵⁸Co activity remained at rather high levels, about 200 GBq/Mg. In units B, C1, C2 (Inconel 690TT RSG), the peak ⁵⁸Co activities reached similar or lower levels than those prior to SGR. In unit B, the peak ⁵⁸Co level is similar, about 100 GBq/Mg 14 cycles after SGR, whereas in unit C1, the peak ⁵⁸Co level is below 15 GBq/Mg from 7 cycles after SGR and even below 1 GBq/Mg from 9 cycles after SGR. As for the surface activities (see §3.1), these different types of behavior are attributed to the SG

tube manufacturing processes which lead to SG tube releases more or less high during operating cycles.

In unit D with Incoloy 800 SG tubes, the peak ^{58}Co activity was relatively high during several cycles after SGR. It was only from cycle 9 after SGR that the peak ^{58}Co levels reached values less than about 5 GBq/Mg, as expected for Incoloy 800 SG tubes.

4. DISCUSSION

Whatever the replacement SG tube alloy used (Inconel 600, Inconel 690 or alloy 800), the ^{58}Co activities deposited inside the primary systems and released during refueling outages generally increase for 1 to 3 cycles after SGR.

This increase in ^{58}Co is mainly due to a high Ni release from the replacement SG tubes during the formation of an inner protective oxide layer on the fresh alloy surfaces. Indeed, no special preconditioning was performed to “passivate” the replacement SG tube surfaces during the initial startup after SGR. However, even if a special preconditioning were performed, its impact could be limited [8] or even negative, because the oxides formed during the Hot Functional Test (HFT) passivation procedure could be thermodynamically unstable in the power operating conditions and subsequently dissolved during the first operating cycle after SGR. Tests performed in the CORELE loop [9] showed this type of behaviour. Nevertheless, an HFT procedure able to form stable oxides in power operating conditions should reduce the Ni release during the first cycles after SGR.

The circuit cleanliness after a SGR process may also have an impact during the first operating cycles following the SGR outage. Indeed, the SGR process could lead to loose oxide detachments or to an addition of metal dust deposited inside the new tubes. Therefore a complete water purification stage after a SGR process or after an HFT phase is recommended [10].

The Ni deposits inside the primary system before SGR should also be considered. During the operating cycles before SGR, Ni released from the Inconel 600MA tubes deposited on the primary surfaces and may dissolve during the cycles after SGR. This “memory” effect decreases with this Nickel reserve.

After the increase in the ^{58}Co surface and volume activities for 1 to 3 cycles following SGR outage, a decrease in the ^{58}Co contamination occurs due to a decrease in the Ni release from the replacement SG tubes. The levels reached after several cycles are mainly due to the manufacturing process of the replacement SG tubes. Indeed, the metal release from SG tubes depends on the SG tubing surface finish related to the manufacturing process [11]. The surface finish is mainly defined by crystallographic, chemical, physicochemical and microgeometric characteristics. At this time, the main parameter controlling the release rate of the corrosion products is not well identified. The levels reached after several cycles do not seem to depend significantly on the SG tube alloy composition because a SGR with alloy 600TT can lead to a ^{58}Co contamination similar to a SGR with alloy 690TT and the ^{58}Co surface activity inside Inconel 690TT SG tubes can be similar to the one inside Incoloy 800 SG tubes.

This is especially true for a same SG tube alloy, such as in units B, C1 and C2 with Inconel 690TT replacement SG tubes: the differences in contamination are due to different SG tube manufacturing processes and thus to different SG tube surface finishes. For instance, even 14 cycles after SGR, peak ^{58}Co activity is about 100 GBq/Mg in unit B, whereas it is 100 times lower in unit C1 from 9 cycles after SGR. The Inconel 690TT SG tube manufacturing process

was probably improved between these two SGRs. The SG tubing release in unit C1 is low enough after the formation of the passive oxide layer so that practically all the nickel forms nickel ferrites. There are practically no metallic Ni deposits on fuel rods anymore from 9 cycles after SGR, and therefore the peak ^{58}Co activity during oxygenation is very low [12].

The SG tube manufacturing process changed again between SGRs in units C1 and C2, but this change was not associated to any further improvement of the SG tube release. Note that the higher levels of the surface and volume activities in unit C2 are not due to the higher replacement SG tube surface area (+14%).

The chemistry conditioning, coordinated chemistry at $\text{pH}_{300^\circ\text{C}} = 7.2$ or 7.0 , is not responsible for the differences or for the variation in the contamination after SGR. The target $\text{pH}_{300^\circ\text{C}}$ was already 7.2 before SGR in unit C2 whereas it was 7.2 after SGR in unit C1 (see §2).

The decrease in the peak ^{58}Co activity during refueling outages in unit C1 after SGR is also observed in the ^{58}Co volume activity during operating cycles as shown in Figure 10. This figure presents the average ^{58}Co volume activity per cycle during operating cycles in units C1 and C2. Even during operating cycles, the level is higher in unit C2 than in unit C1 from the third cycle after SGR.

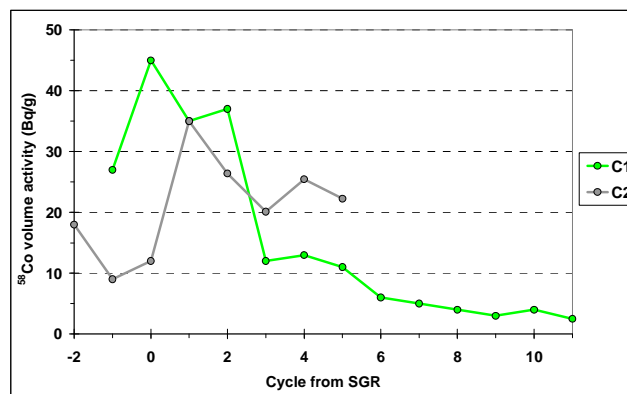


Figure 10. Average ^{58}Co volume activity during operating cycles.

The SG tube manufacturing process also has an impact on the variation in the ^{60}Co surface activity inside the primary system after SGR. Due to a generally lower Co content of the replacement SG tubes and to the improvement in their manufacturing process, the ^{60}Co surface activity increases by a lower rate than before SGR inside the new SG tubes and tends to decrease inside the primary coolant pipes. However, due to the long ^{60}Co radioactive half-life (5.3 years), the contamination levels after SGR mainly result from the actual state existing before SGR.

The variations in the ^{58}Co and ^{60}Co contaminations after SGR lead to an increase in the ^{58}Co contribution to the dose rates around the primary system. ^{58}Co generally becomes the major contributor to dose rates for the first cycles after SGR then its contribution progressively decreases giving way to ^{60}Co because, on the one hand, the ^{58}Co surface activity decreases and, on the other, ^{60}Co accumulates due to its longer radioactive half-life.

As the steam generator tubes represent the largest primary surface area (about 64% for a 900 MWe PWR), after commissioning and after SGR there is a similitude in the variation in the

^{58}Co and ^{60}Co total activities deposited on the primary out-of-core surfaces as shown for unit B in Figure 11.

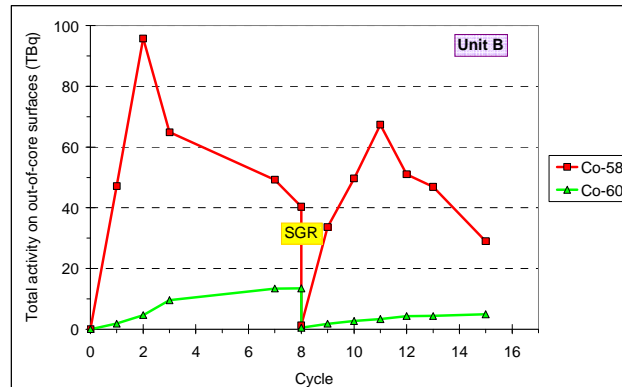


Figure 11. ^{58}Co and ^{60}Co total activities deposited on the primary out-of-core surfaces in unit B.

5. CONCLUSION

Given the fact that the steam generator tubes represent the largest primary surface area, a steam generator replacement may greatly impact the primary system contamination. After SGR, whatever the SG tube material is, the typical variations are the following:

- The ^{58}Co activities deposited inside the primary system and released during refueling outage increase for 1 to 3 cycles and then decrease to very low levels in some cases mainly depending on the material properties of the replacement SG tubes.
- The ^{60}Co surface activity tends to decrease on the primary coolant pipes and to increase by a lower rate on the new SG tubes.

This analysis, based on experimental feedback enriched over the past 10 years, confirms the conclusion of the previous study [1]: the two main key factors in the contamination levels after SGR seem to be the *corrosion product deposits prior to SGR* (especially for the ^{60}Co contamination) and the *surface finish of the replacement SG tubes related to their manufacturing process* (impact on the SG tube release and deposit). The SG tube alloy (alloys 600, 690, 800) seems to be of secondary importance.

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