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EMECC CAMPAIGN AT TRILLO : RESULTS AND COMPARISON WITH FRENCH FEEDBACK

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ABSTRACT

The reduction of collective dose is an important objective for all nuclear plant operators. One of the support for studying the impact of design and operation parameters is the measurement campaigns of contamination in PWRs. EDF performs consequently gamma spectrometry measurements with CEA and his EMECC system, with more than 300 campaigns in EDF plants and also some campaigns in collaboration with other operators.

Commissioned in 1988, the only unit of the nuclear power plant of TRILLO, located in Spain, is a 3-loop PWR of 1066 MWe designed by KWU-Siemens. TRILLO-1, equipped with alloy 800 tubed steam generators, is characterized by a low dosimetry. In order to compare the contamination level of TRILLO-1 to that of French PWRs, an EMECC campaign was performed within the framework of a contract between EDF and the CEA, and with the agreement of CNAT/TRILLO and the support of the CSN (Consejo de Seguridad Nuclear).

This campaign took place during the refueling shutdown at the end of cycle 21 from 9 to 16 February 2009. A previous EMECC campaign was performed in 1992 during the refueling shutdown of cycle 4.

At the first time, this paper will present the main conclusions of this EMECC campaign :

- deposited activities measured on legs walls and steam generator tubes walls,
- dose rates measurements.

The evolution of the contamination since cycle 4 will be also analyzed.

Furthermore, a comparison with the French unit St-LAURENT B1 will be achieved in order to better understand the origin of the differences (source term or outage management) and to improve good operational practices in the future.

1. INTRODUCTION

Commissioned in 1988, the only unit of the nuclear power plant of TRILLO, located in Spain, is a 3-loop PWR of 1066 MWe designed by KWU-Siemens. TRILLO-1 equipped with alloy 800 tubed steam generators, is characterized by a low dosimetry (382 Man.mSv in 2008).

In order to compare the contamination level of TRILLO-1 to that of the French unit St-LAURENT B1, an EMECC campaign [1.] was carried out within the framework of a contract

between EDF and CEA and with the agreement of CNAT/TRILLO and the support of the CSN (Consejo de Seguridad Nuclear i.e. the Spanish Nuclear Safety Authority).

This campaign took place during the refueling shutdown at the end of cycle 21 from February 9th to 16th 2009. A previous EMECC campaign was conducted in 1992 during the refueling shutdown of cycle 4.

1. OPERATIONNAL PARAMETERS AT TRILLO

1.1 Reactor coolant chemistry

In TRILLO-1 during cycle 21 (281 EFPD), the reactor coolant was based on a boron-lithium coordinated chemistry with a $\text{pH}_{300^\circ\text{C}}$ target of 7.5. Almost immediately at the beginning of cycle 21, $\text{pH}_{300^\circ\text{C}}$ rose above 6.9 whereas the maximum Li content has remained quasi-stable at about 2.2 ppm for 80% of the cycle duration. This procedure is slightly different than the French one for which the $\text{pH}_{300^\circ\text{C}}$ target is 7.2 (optimum chosen by EDF between contamination risk and potential PWSCC long term risk with high lithium concentration).

During cycle 21, H_2 concentration in primary coolant ranged between 22 and 28 cm^3/kg (TPS), with an average of 24 cm^3/kg .

1.2 Cold shutdown procedure

Various shutdown parameters are shown in Figure 1 (temperature, letdown flow-rate, boron, hydrogen and oxygen concentrations and ^{58}Co volume activity).

Considering the very low activity level of ^{58}Co (1.33 GBq/t), the last RCP pump shutdown occurred only 2.5 hours after RHRS connection.

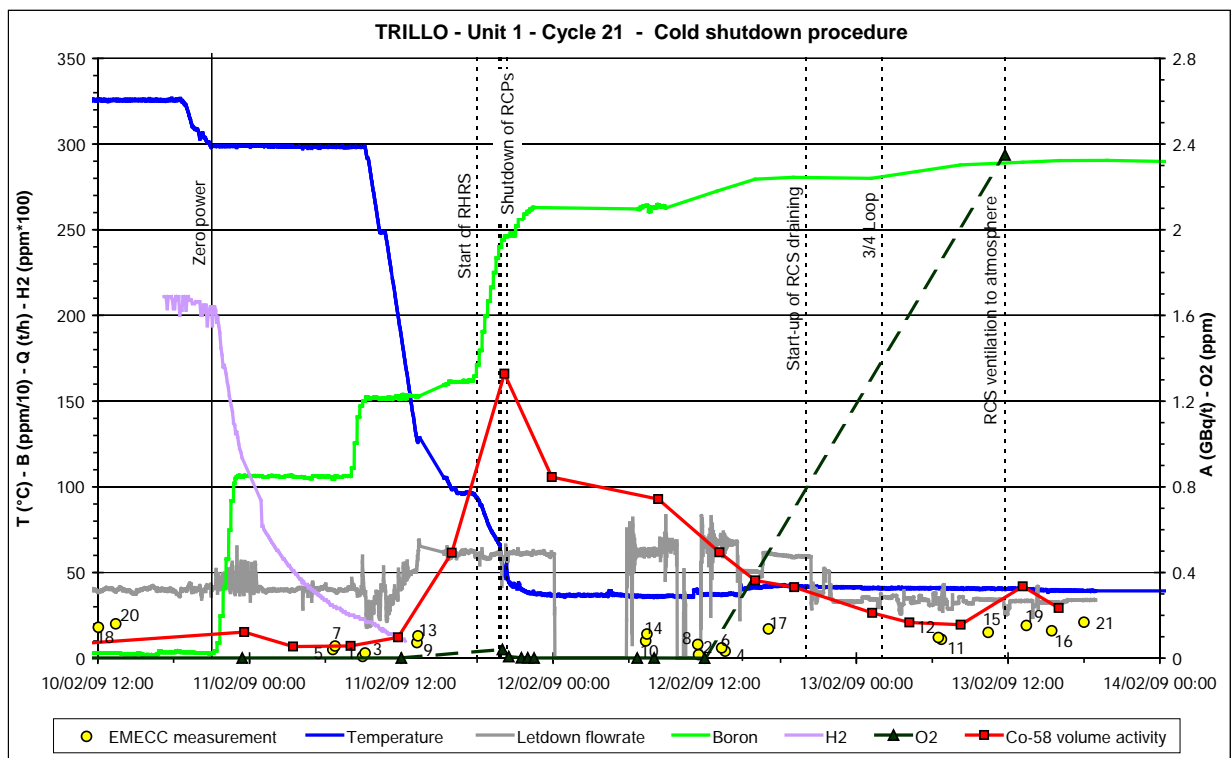


Figure 1. TRILLO 1 Cycle 1 - Cold shutdown procedure

2. EMECC CAMPAIGN RESULTS AT TRILLO-1 AND COMPARISON WITH SAINT-LAURENT B1

2.1 Volume activities

During cycle 21

A comparison of volume activities during TRILLO 1 cycle 21 and St LAURENT B1 is shown in table 1.

Table 1. Comparison CNT121/SLB120 – Average volume activities during cycle

MBq/t	CNT 121	SLB 120
⁵⁸ Co	1.8	3.1
⁶⁰ Co	1.2	1.8
⁵⁴ Mn	0.9	1.5
⁵¹ Cr	6.4	8.9

One can observe that the volume activities in CNT121 are slightly lower than those measured in SLB1.

During shutdown

RCS oxygenation is the most important stage of the French cold shutdown procedure because it usually causes a very large increase in the ⁵⁸Co volume activity (order of magnitude of 100 GBq/t), which can lead to an increase in the activities deposited on the out of flux surfaces [8.]. To control this stage, H₂O₂ is injected when the primary coolant temperature is 80°C. Nevertheless, due to an improved manufacturing process of the alloy 690 TT SG tubes (Ni : 60%; Fe : 10%; Cr : 30%) of St-LAURENT B1 [4], the ⁵⁸Co peak is very low at cycle 20 : 0.8 GBq/t.

In TRILLO 1, thanks to the alloy 800 SG tubes (Ni : 35%; Fe : 45%; Cr : 20%), the primary system oxygenation leads to a very low ⁵⁸Co peak (order of magnitude of 1 GBq/t) typical of Konvoy PWRs, therefore there is less need to control it. At cycle 21, the primary system oxygenation occurred during the start of RHRS, perhaps during the draining of RCS (increase of ^{110m}Ag volume activities) and of course during the primary system ventilation to atmosphere.

The ⁵⁸Co peaks in PWRs equipped with alloy 690 SG tubing, like St-LAURENT B1, can be as low as those of the Konvoy PWRs thanks to a very low SG tubing release. Therefore, more than the material (alloy 690 or alloy 800), the key factor leading to very low ⁵⁸Co peaks is the SG tubing surface state obtained by means of an improved tubing manufactured process.

Moreover, in TRILLO 1, the low ⁵⁸Co peak could be explained by the fact that practically all the nickel released by the alloy 800 SG tubes generates nickel ferrites. Thus, there is almost no Ni⁰ or NiO deposited on the fuel elements and potentially releasable during the oxygenation peak. Nickel ferrites are more stable in oxidizing medium and their solubility increases with temperature in optimum conditions (no preferential deposits on fuel elements). In St-LAURENT B1, the lack of Ni⁰ or NiO is due to a very low release of alloy 690 SG tubing [7.].

2.2 Surface activities

Average surface activities measured at TRILLO 1 cycle 21 are presented in table 2 for legs, SG tubes and CVCS non regenerative heat exchanger (NRHE).

Table 2. Average surface activities for some radionuclides in TRILLO 1

GBq/m ²	⁵⁸ Co	⁶⁰ Co	⁵⁴ Mn	¹²⁴ Sb	^{110m} Ag	⁵⁸ Co/ ⁶⁰ Co ratio
Legs	0.76	2.69	0.16	0.08		0.28
SG tubes	0.2	0.49		0.07		0.4
CVCS letdown line (NRHE)	0.004	0.006		0.004	0.022	0.66

Average surface activities measured at St LAURENT B1 cycle 20 are presented in table 3 for legs, SG tubes and CVCS letdown line.

Table 3. Average surface activities for some radionuclides in St LAURENT B1

GBq/m ²	⁵⁸ Co	⁶⁰ Co	⁵⁴ Mn	¹²⁴ Sb	^{110m} Ag	⁵⁸ Co/ ⁶⁰ Co ratio
Legs	1.52	1.34	0.21	0.21	0.15	1.1
SG tubes	0.53	0.29	0.04	0.07		1.8
CVCS letdown line (NRHE)	0.04	0.04	0.006	0.047	0.32	1.0

At TRILLO 1, one can observe that the main contributors to activity are ⁵⁸Co ($\approx 20\%$) and ⁶⁰Co ($\approx 65\%$) in RCS. The same radionuclides also mainly contribute to surface activity at St-LAURENT B1 but the ⁵⁸Co impact is significantly higher (40%-60% for ⁵⁸Co and $\approx 35\%$ for ⁶⁰Co) as shown by the different ratios ⁵⁸Co/⁶⁰Co.

One can also notice a specific contribution of ^{110m}Ag ($\approx 50\text{-}60\%$) in CVCS non regenerative heat exchanger. A same tendency is observed at St-LAURENT B1. This phenomenon, well-known in French PWRs [2.], can be explained by thermochemical considerations. Silver is soluble in oxidizing conditions and its solubility increases with temperature. Thus, during oxygenation, it dissolves in the primary coolant from RCS deposits and then precipitates on the cold parts of auxiliary systems, in particular heat exchangers.

Deposited activities in ⁵⁸Co and ⁶⁰Co are presented in figure 2 for hot legs, in figure 3 for crossover legs, in figure 4 for SG tubes and in figure 5 for CVCS non-regenerative heat exchanger.

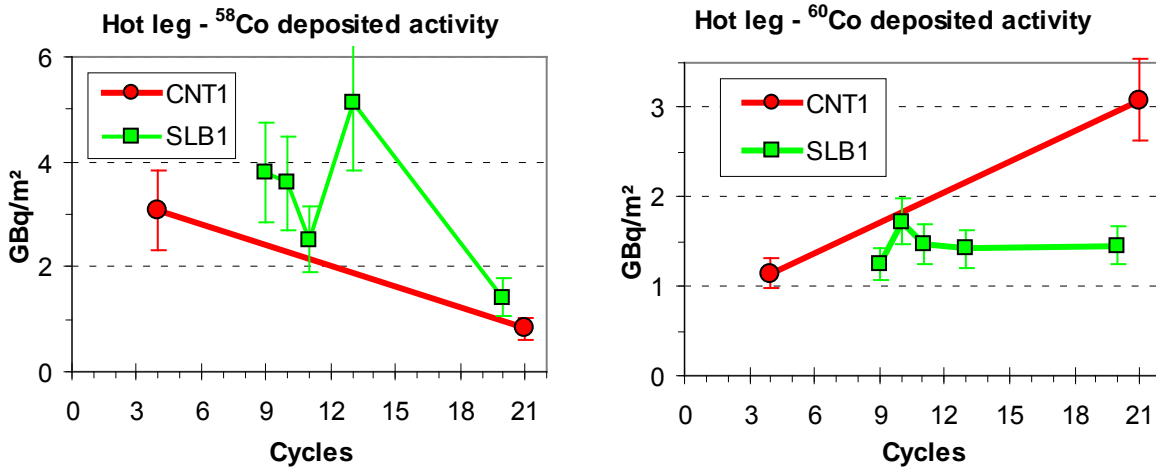


Figure 2. Comparison CNT121/SLB120 - ⁵⁸Co and ⁶⁰Co surface activity on Hot Legs

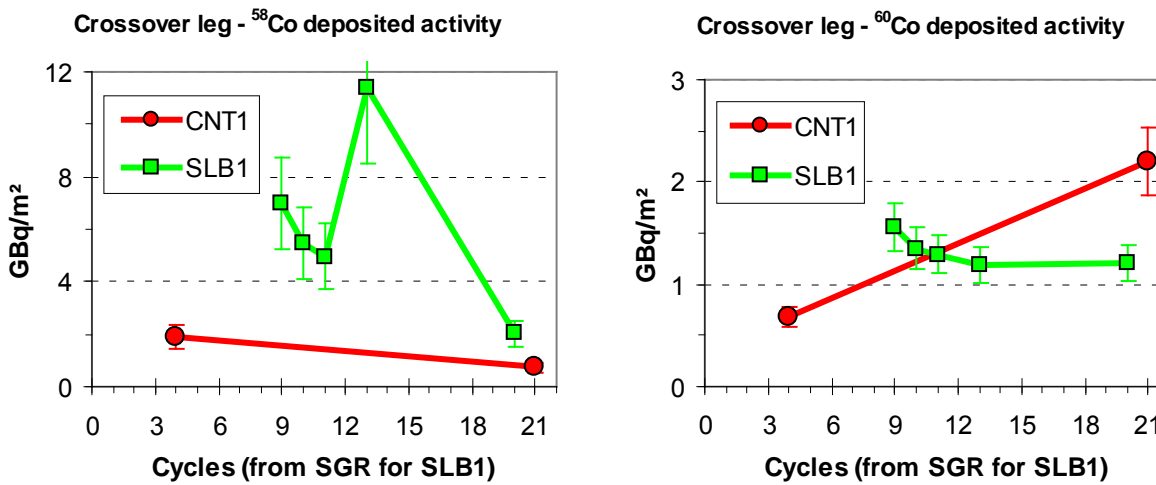


Figure 3. Comparison CNT121/SLB120 - ⁵⁸Co and ⁶⁰Co surface activity on Crossover Legs

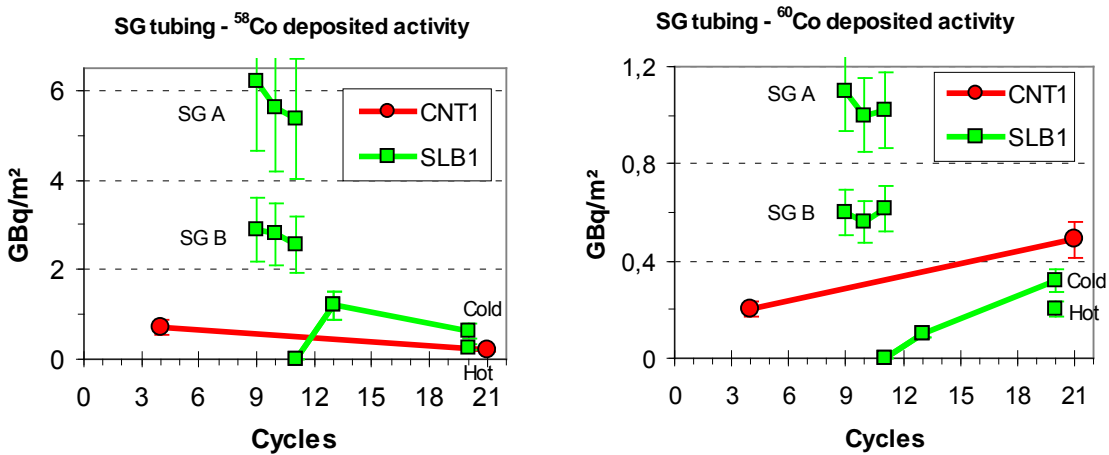


Figure 4. Comparison CNT121/SLB120 - ⁵⁸Co and ⁶⁰Co surface activity on SG tubing

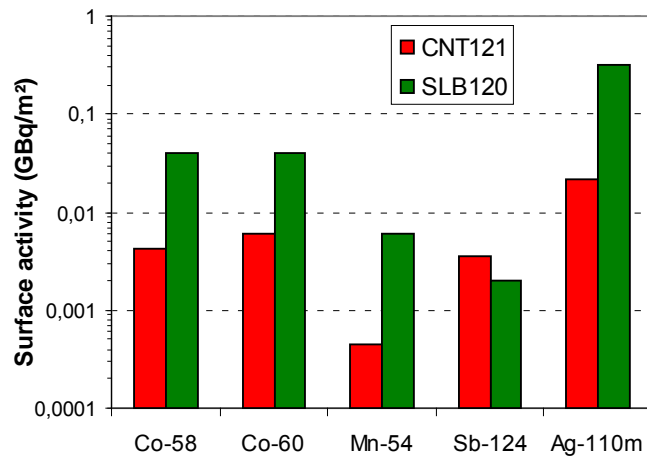


Figure 5. CVCS letdown pipe surface activity – Comparison Trillo 1 / Saint Laurent B1

There is no difference in contamination level between SG tubes hot and cold sides at TRILLO 1 whereas the SG tube cold side at St LAURENT is about 2 times higher than the hot side. This phenomenon has rarely been pointed out on the French fleet and has been mainly observed after a steam generator replacement [3.]. This could be explained by a probable strong dependence of the new SG tube oxides solubility to temperature due to differences into stoichiometric ratios between old and new SG tubes.

On one hand, ⁵⁸Co surface activity at TRILLO 1 has decreased by a factor 3.8 on average inside primary system since cycle 4. On the other hand, ⁶⁰Co surface activity has increased by a factor 2.6 on average. These tendencies are consistent with those generally observed in French PWR [4.].

2.3 Dose rates

Dose rate measurements were performed around the RCS in order to estimate CEA indexes (figure 8) on primary piping and on SG tubes bundle. The results are reported in table 4.

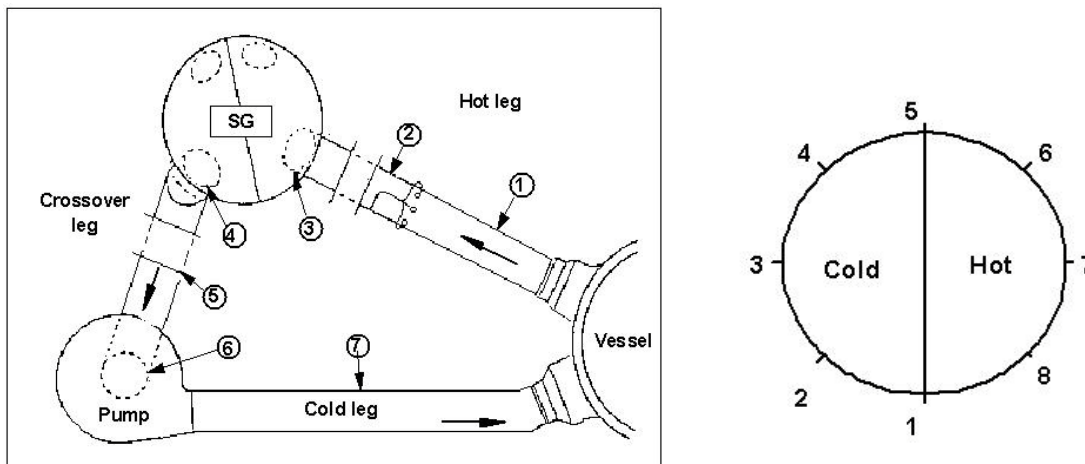


Figure 8. Primary piping and SG tube bundle : CEA dose index

Table 4. Comparison CNT121/SLB120 – CEA dose rate index

mSv/h	CNT 121	SLB120	CNT121 / SLB120 Ratio
Primary piping	0.49	0.27	1.8
SG tube bundle	0.02	0.09	0.2
CVCS NRHE	0.23	2.3	0.1

Primary piping :

The average dose rate measured around primary system is higher in TRILLO 1 Cycle 21 than at St LAURENT B1 Cycle 20. The higher dose rate index of TRILLO primary piping is due to higher ^{60}Co surface activities (factor 2) and to a lower wall thickness (about 6 cm at TRILLO compared to about 7 cm at St-LAURENT). However, one can notice that the dose rate index at TRILLO would have been similar to St-LAURENT if the component thickness had been identical in spite of a higher level of ^{60}Co contamination because the ambient dose rate was lower at TRILLO than at St-LAURENT (contaminated by-pass pipes influence).

SG Tubes :

The lower dose rate index of the TRILLO SG tube bundle is due to a higher SG shell thickness (10 cm in TRILLO compared to 8.5 cm in St-LAURENT). The dose rate index of TRILLO would have been similar to the St-LAURENT one if the component thickness was identical.

CVCS Regenerative Heat Exchanger :

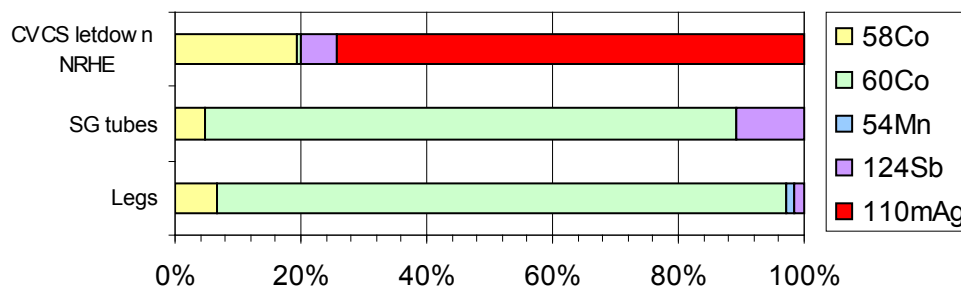
The dose rate close to the non regenerative heat exchanger is 10 times lower in TRILLO 1 than in St-LAURENT B1 : 0.23 mSv/h compared to 2.3 mSv/h in SLB120. This is essentially due to the contamination caused by $^{110\text{m}}\text{Ag}$ which is the major contributor (about 65% to 80%) whereas ^{60}Co contribute for only 15% to 20%.

Contributors to dose rate :

As shown in figures 9 and 10, ^{60}Co is proved to be the major contributor to the dose rates around the primary system at CNT121 (between 80% and 90%), and SLB120 (between 65% and 75%).

Contribution from ^{58}Co was about 5% in CNT121 and ranged between 10% and 25% in SLB120.

Around SGs, ^{124}Sb the contribution is not higher than 10%.

**Figure 9. TRILLO 1 Cycle 21 – Radionuclide contribution to dose rates**

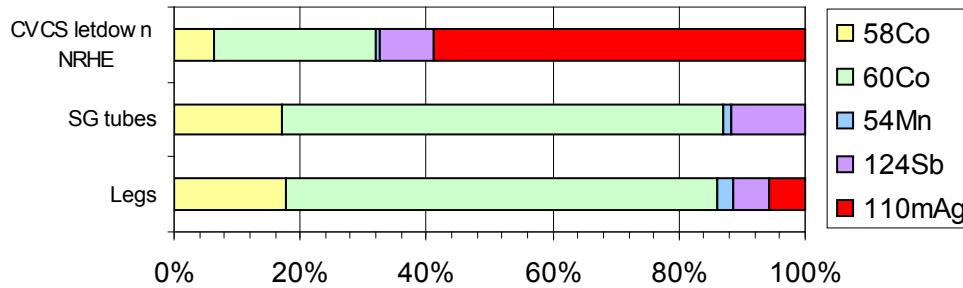


Figure 10. St-LAURENT B1 Cycle 20 – Radionuclide contribution to dose rates

2.4 Collective dosimetry

The comparison between TRILLO 1 and St-LAURENT B1 indicates that TRILLO 1 had a very lower collective dosimetry than St-LAURENT B1 until 1999. This is due on one hand to higher contamination levels in ⁵⁸Co and ⁶⁰Co at St-LAURENT B1 mainly on SG tubes (see figure 2 to 5) and on the other hand probably to outage management. St-LAURENT B1 staff have performed a significant improvement program for the last decade, due to efficient ALARA plans. As a result of these actions, figure 9 shows quite similar collective dosimetry for both units in the last 3 years.

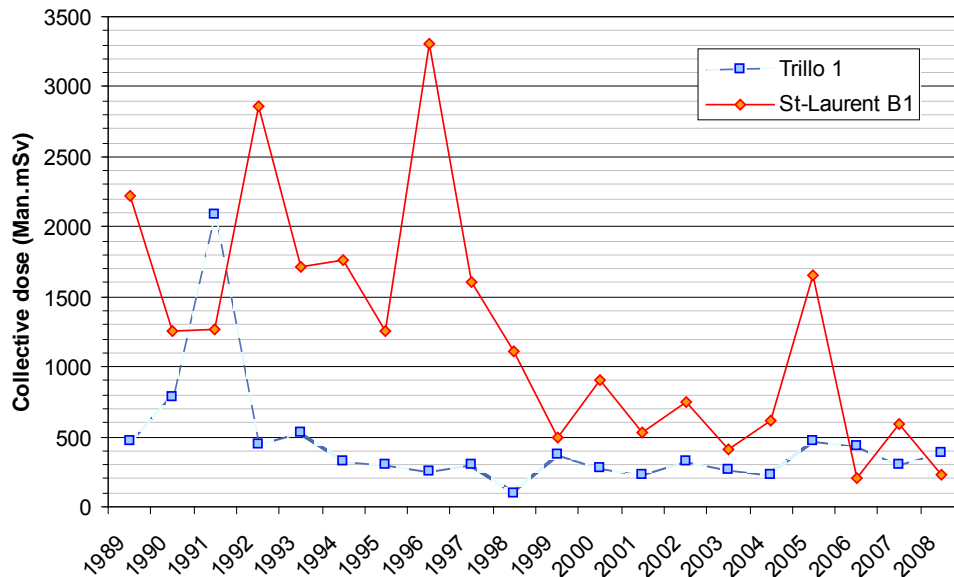


Figure 9 : Collective dose in TRILLO-1 and St-LAURENT B1 from 1989 to 2008

3. DISCUSSION

The ⁶⁰Co surface activity inside the primary system of TRILLO 1 at cycle 21 was higher than that of St-LAURENT B1 at cycle 20 and than that of Konvoy PWRs. The possible reasons for this higher ⁶⁰Co contamination level in TRILLO 1 are examined in this section hereafter.

First of all, a chemical conditioning such as the one at cycle 21 can not be the cause of this specific high ^{60}Co contamination level. In effect, it has been shown that primary chemistry may have a potential positive (and not negative) impact on contamination [5.] and there is no reason for the impact to only affect ^{60}Co .

Stellite hard-facing alloys could be responsible of this elevated ^{60}Co source term in case of abnormal wear, for instance during the grinding of valve seats. An abnormal wear of a component such as pump bearings (made of stainless steel with increased Co content) could also be a possible origin of contamination. However, an abnormal wear is generally characterized by the presence of hot spots inside the circuits, which is not the case at TRILLO 1.

Furthermore, Inconel 718 fuel grids could be the main cause of that observed ^{60}Co contamination. In fact, since the beginning, fuels assemblies at TRILLO 1 have always contained Inconel 718 grids. During the four first cycles, all fuel spacer grids was made of Inconel 718. Afterwards and until cycle 17, only bottom and top grids were made of Inconel 718, the others grids being made in Zircaloy-4. More recently, the refueling assemblies have been made with a bottom fuel grid made of Inconel 718 and the others made of HPA-4. Since the Co specification of alloy 718 grids was high (1000 ppm), it is possible that the grids (activated by intense neutronic flux) have directly released Co into the primary coolant leading to a ^{60}Co contamination build-up.

It should also be pointed out that ^{60}Co and especially ^{58}Co surface activities were already relatively high at the end of cycle 4. Thus, this contamination, that probably occurred during the first cycles, could involve a nickel-based material as Inconel 718 grids (about 52% of Ni).

Moreover, the nickel concentration has been high in operation since cycle 16 at least (between 1 and 10 ppb) and do not decrease as cycles go along. This level is not common, particularly in a PWR equipped with alloy 800 SG tubes (the usual values are measured between 0.1 and 1 ppb [9.]) and seems to show that Ni source term is “regenerated” after each outage, what could be consistent with refueling assemblies equipped with Inconel 718 grids.

Contamination levels at TRILLO-1 look, to a lesser extent, like those of BIBLIS-B, a pre-KONVOY PWR *i.e.* low ^{58}Co contamination levels (surface and released) and relatively high ^{60}Co surface activity for TRILLO-1 or very high for BIBLIS-B. The very high ^{60}Co contamination level in BIBLIS-B is due to the use of nickel-plated fuel spacers with an increased cobalt content [6.]. Inconel 718 fuel grids could also be the cause in TRILLO-1.

A priori, the SG tubing Co content is not responsible for this high ^{60}Co source term : the specified mean value was 150 ppm in TRILLO, which is similar to St-LAURENT-B1(measured content of 122 ppm) [4.].

Finally, The fact that the Ni concentration has remained at a high level for several cycles could be also due to the CVCS design : there is no particle filter upstream from the ion exchange resins. It seems that neither the mixed bed resins nor the mechanical filter downstream from them could trap particles efficiently.

4. CONCLUSION

The EMECC campaign performed during the beginning of the refuelling outage planned at the end of cycle 21 in TRILLO-1 revealed the following points in particular :

- The ^{58}Co surface activities on the primary coolant pipes and on the steam generator tubing in TRILLO-1 at cycle 21 were 2 times lower than those in St-LAURENT-B1 at cycle 20 and similar to those in the Konvoy PWRs. On the other hand, the Ni concentration has been abnormally high for several cycles in TRILLO-1.
- The ^{58}Co peak during the outage 21 in TRILLO-1 was very low and similar to the one in SAINT-LAURENT-B1 at cycle 20 and to those in the Konvoy PWRs.
- The reason for the low ^{58}Co surface activities and ^{58}Co peak in TRILLO-1 is the SG tubing alloy 800 (low Ni content), whereas the low ^{58}Co peak in St-LAURENT-B1 at cycle 20 is due to the alloy 690 SG tubing surface finish.
- The ^{60}Co surface activities of the primary system were higher by a factor 2 in TRILLO-1 at cycle 21 compared to SAINT-LAURENT-B1 at cycle 20. The use of Inconel 718 fuel grids with a high Co impurity content must have played a great part in this high ^{60}Co contamination level.
- The main contributor to the dose rates around the primary circuit in TRILLO-1 at cycle 21 was ^{60}Co , between 80% and 90%. As the ^{60}Co radioactive half-life is 5.3 years, the dose rates will remain relatively high for many cycles to come.

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