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## **Influence of the dissolved hydrogen concentration on the contamination of the primary loop of DOEL PWR using the OSCAR code**

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### **Abstract**

Normal operation of PWR generates corrosion products in the primary circuit which are activated in the core and constitute the major source of the radiation field.

To minimize these risks and constraints, it is essential to understand the behavior of corrosion products by carrying out the appropriate measurements in PWR circuits and loop experiments combined with reactor contamination assessment code.

The reactor contamination assessment code allows to take into account the chemical and physical mechanisms in different situations in operating reactors or at design stage. OSCAR code has been developed with this aim by CEA in collaboration with EDF and Framatome, and has actually been used since the early seventies [1].

OSCAR is a reliable tool for PWR (also used for EPR, SFR, ITER, decommissioning...) calibrated and validated with a complete database of contamination measurements in EDF water reactors.

Water chemistry has an influence on corrosion of the main materials (especially nickel-based alloys), in Belgian PWR the average dihydrogen concentration used is around 30 cc/kg, which is in line with current international standards however which is not the best value to reduce stress corrosion cracking of the materials. It also has an influence on dissolution/precipitation mechanisms involved in contamination.

Water chemistry control allows then to reduce significantly the radioactive contamination in the primary loop and therefore facilitates maintenance operations.

In this field, hydrogen plays a critical role in limiting the presence of oxidizing species due to water radiolysis. Increasing hydrogen could also reduce core internals cracking.

The aim of this study is to evaluate the influence of an increase/decrease of the Dissolved Hydrogen (DH) concentration on the contamination of the primary loop using the OSCAR code.

This study presents the simulation results of a sensitivity analysis, using the 1.3 version of the OSCAR code, of the contamination in the primary loop of DOEL PWR with DH concentrations ranging between 15 and 70 mL/kg.

The evolution of the surface contamination in  $^{58}\text{Co}$  and  $^{60}\text{Co}$  were calculated on the hot legs, U tubes and Steam Generators (SG) tubing along with the mass of Ni deposited on the fuel. In order to explain those evolutions, equilibrium (concentration of the element in the coolant with respect to the considered oxide) and ionic Ni concentrations were investigated on the SG and the fuel as well as the Ni dissolution and release flux of the SG.

## **Introduction**

Normal operation of PWR generates corrosion products in the primary circuit, which are activated in the core and constitute the major source of the radiation field.

To minimize these risks and constraints, it is essential to understand the behavior of corrosion products by carrying out the appropriate measurements in PWR circuits and loop experiments combined with reactor contamination assessment code.

The reactor contamination assessment code allows to take into account the chemical and physical mechanisms in different situations in operating reactors or at design stage. The OSCAR code [2] has been developed with this aim by CEA in collaboration with EDF and Framatome, and has actually been used since the early seventies.

OSCAR is a reliable tool for PWR (also used for EPR, SFR, ITER, decommissioning...) calibrated and validated with a complete database of contamination measurements in EDF water reactors.

Water chemistry has an influence on corrosion of the main materials (especially nickel-based alloys), in the Belgian PWRs the average dihydrogen concentration used is around 30 cc/kg, which is in line with current international standards however which is not the best value to reduce stress corrosion cracking of the materials. It also has an influence on dissolution/precipitation mechanisms involved in contamination. Water chemistry control then allows to reduce significantly the radioactive contamination in the primary loop and therefore facilitates maintenance operations.

In this field, hydrogen plays a critical role in limiting the presence of oxidizing species due to water radiolysis. Increasing hydrogen could also reduce core internals cracking.

The aim of this study is to evaluate the influence of the change in the Dissolved Hydrogen (DH) concentration on the contamination of the primary loop using the OSCAR code.

This study presents the simulation results of a sensitivity analysis, using the 1.3 version of the OSCAR code, of the contamination in the primary loop of DOEL PWR with DH concentrations ranging between 15 and 70 mL/kg. It also presents autoclave experiments to evaluate the impact of several DH concentrations on INCOLOY 690 material, which are compared to simulation results.

## **Operating Parameters of DOEL-4**

DOEL-4 is a 3-loop PWR equipped with INCOLOY 690 tubed steam generators since the steam generator replacement at cycle 11.

The structure of the DOEL-4 primary circuit has been modelled by control volumes using the design data of the reactor (wetted surfaces, hydraulic diameters, material compositions, nominal temperatures ...).

Operating cycles of DOEL-4 were simulated using the 1.3 version of the OSCAR code with the real hydrogen concentrations for cycles 1 to 22. Three more reference cycles were added to DOEL-4 to try various hydrogen concentrations [3].

The parameters of the reference cycles are reported in Table 1.

Table 1: Parameters of the reference cycles

Nominal Power	Cycle duration	B <sub>init</sub>	Li <sub>init</sub>	DH	pH
100%	473 days	1424 ppm	3.35 ppm	15 – 30 – 70 mL/kg	7.2 (at 312 °C)

The operating parameters (Power, C<sub>B</sub>, C<sub>Li</sub>, C<sub>H2</sub>, C<sub>O2</sub>) are given in Figure 1.

The last three cycles correspond to the reference cycles with a simulated shutdown at the end of each cycle.

Power is set to 100%, the boron concentration decreases at each cycle from 1424 pm to 157 ppm, the lithium concentration decreases from 3.35 ppm to 0.59 ppm. Concerning hydrogen, three cases are studied 15, 30 and 70 mL/kg with a stable value during the three cycles for each case. The oxygen peaks observed are due to the shutdowns.

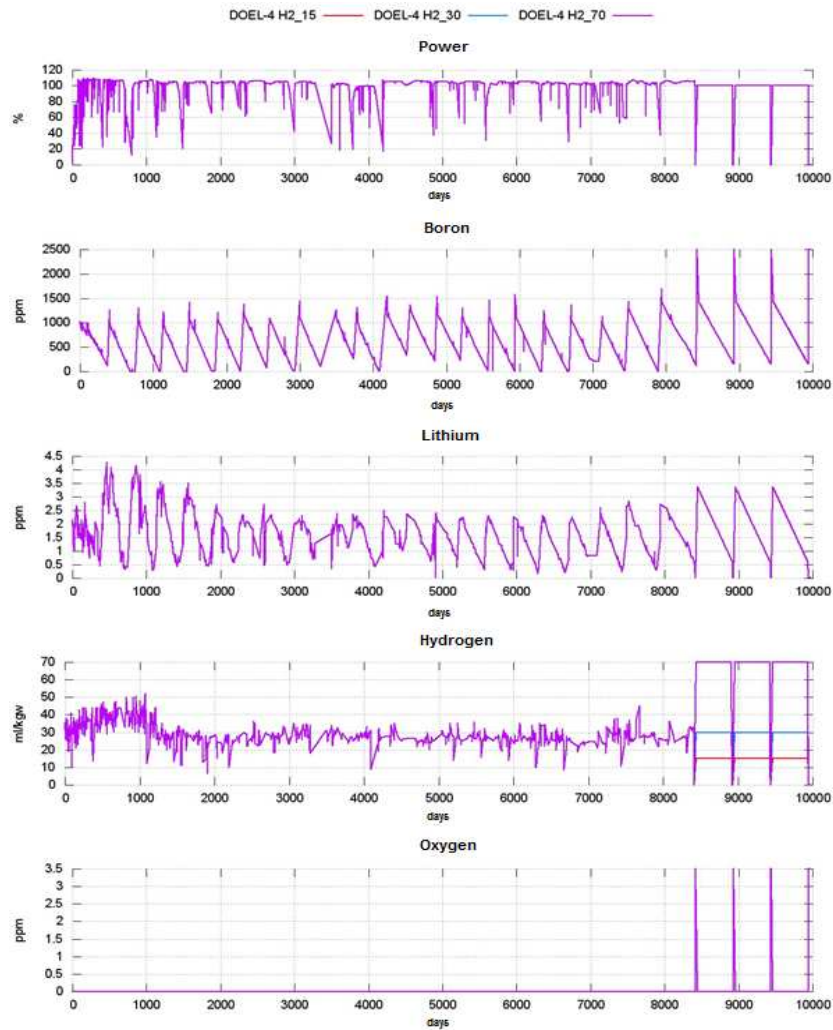


Figure 1: Operating parameters

## Influence of the DH Concentration on the Contamination of DOEL-4 PWR

### Surface activities

$^{58}\text{Co}$  and  $^{60}\text{Co}$  surface activities inside the primary system (Hot leg, Crossover leg, hot side of the Steam Generator tubing, cold side of the Steam Generator tubing) are presented from Figure 2 to Figure 5.

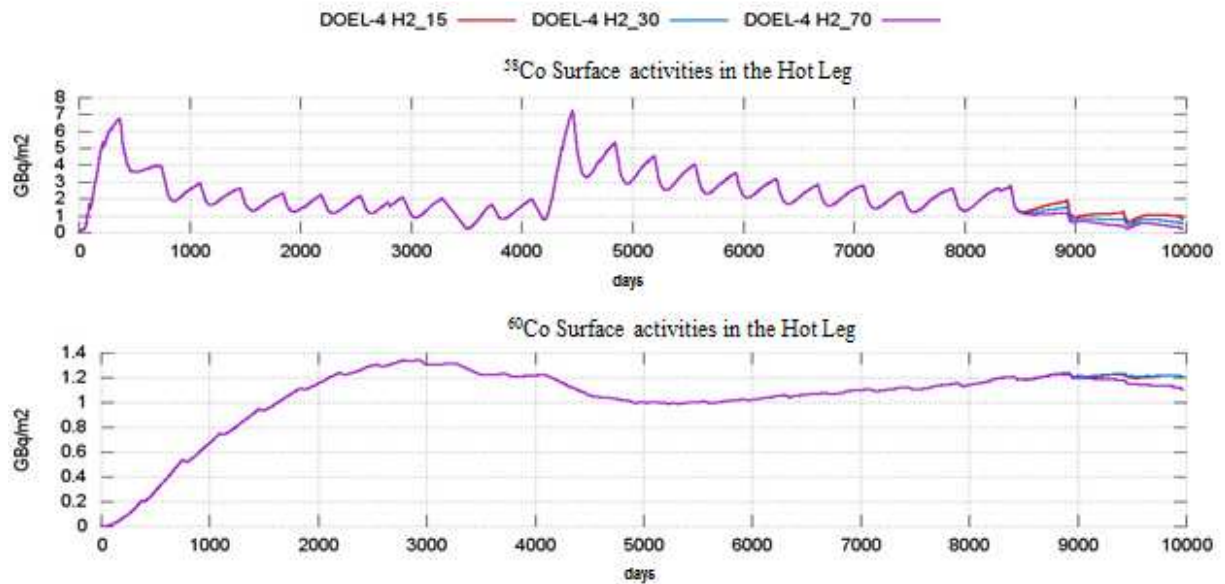


Figure 2: Surface activities on the Hot Leg

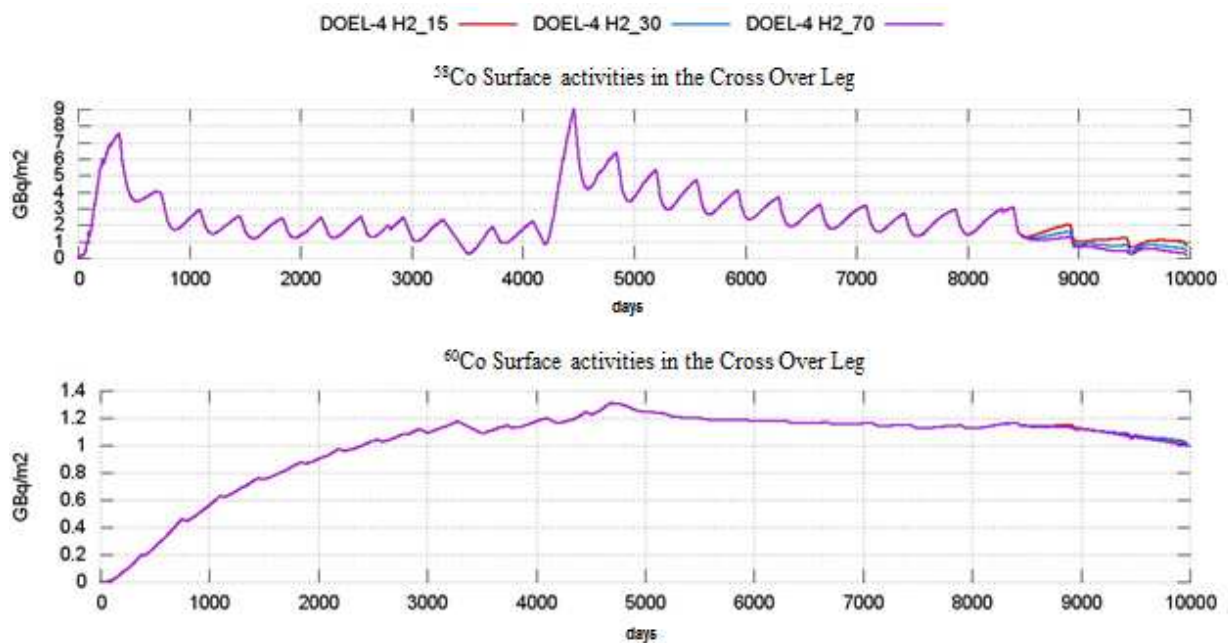


Figure 3: Surface activities on the Crossover Leg

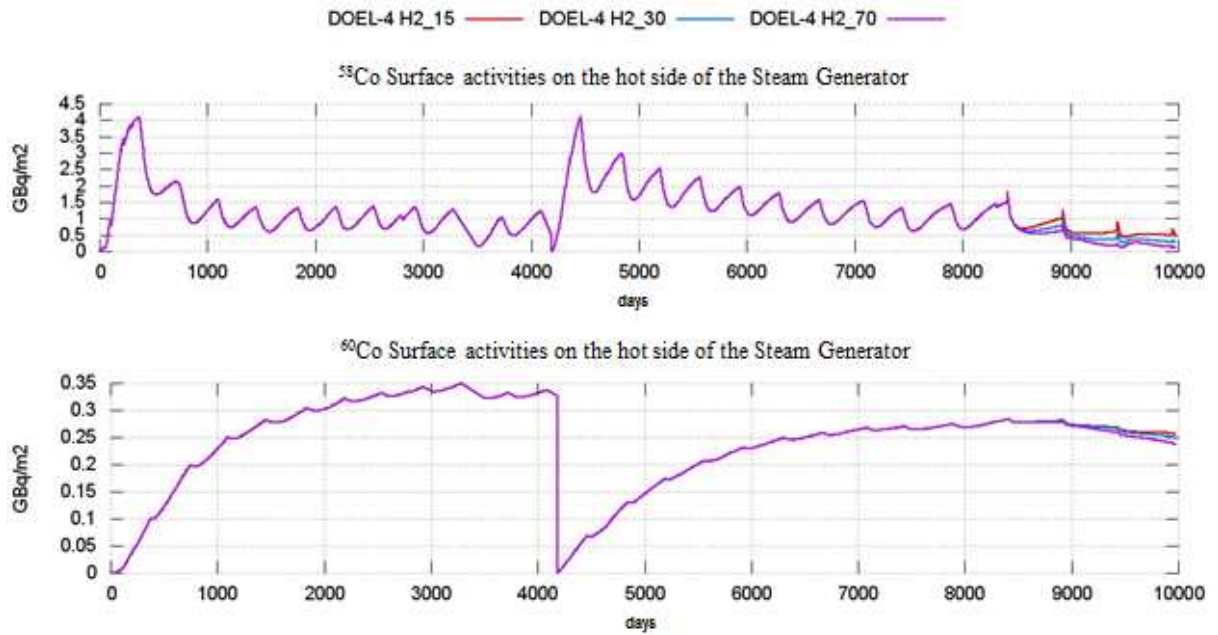


Figure 4: Surface activities on the hot side of the Steam Generator tubing

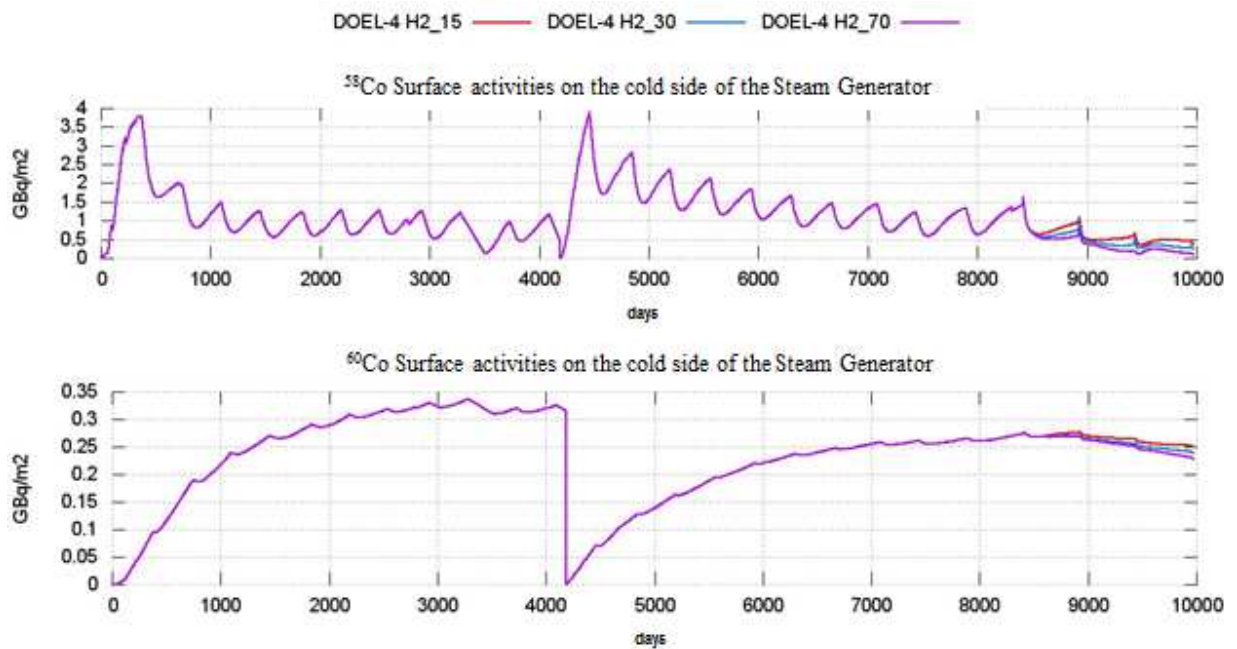


Figure 5: Surface activities on the cold side of the Steam Generator tubing

Concerning  $^{58}\text{Co}$ , it is clear that an increase of the DH value leads to a decrease in the surface activities in the primary system on the last three cycles of DOEL-4. Concerning  $^{60}\text{Co}$ , the decrease tendency is slight.



For DH concentrations of 15 and 70 mL/kg, the relative variations of the total activities (of  $^{58}\text{Co}$  and  $^{60}\text{Co}$ ) deposited on the out of core surfaces at the end of cycle 25 compared to a DH value of 30 mL/kg are presented in Table 2.

The increase in the DH concentration up to 70 mL/kg leads to a decrease in the total activities of  $^{58}\text{Co}$  and  $^{60}\text{Co}$  respectively by 57% and 4%.

On the contrary, a decrease in the DH down to 15 mL/kg leads to an increase of the total activities of  $^{58}\text{Co}$  and  $^{60}\text{Co}$  respectively by 74% and 4%.

Table 2: Relative variations of the total activities deposited on the out of core surfaces compared to a DH value of 30 mL/kg

Variations / [H <sub>2</sub> ] = 30 mL/kg Cycle 25	Out of core surface activities	
	$^{58}\text{Co}$	$^{60}\text{Co}$
[H <sub>2</sub> ] = 15 mL/kg	+74%	+4%
[H <sub>2</sub> ] = 70 mL/kg	-57%	-4%

This table shows that DH concentration significantly affects the  $^{58}\text{Co}$  contamination of the out of core surfaces and to a negligible extent the  $^{60}\text{Co}$  contamination.

It also shows that an increase in the DH concentration (from 15 to 70 mL/kg) lead to a decrease of the total activities of  $^{58}\text{Co}$  and  $^{60}\text{Co}$ .

Ni and Fe masses deposited on the fuel rods are presented in Figure 6. The Ni mass deposited on the fuel decreases when the DH concentration increases (same evolution as the surface activities). On the contrary the Fe mass deposited on the fuel increases when the DH concentration increases.

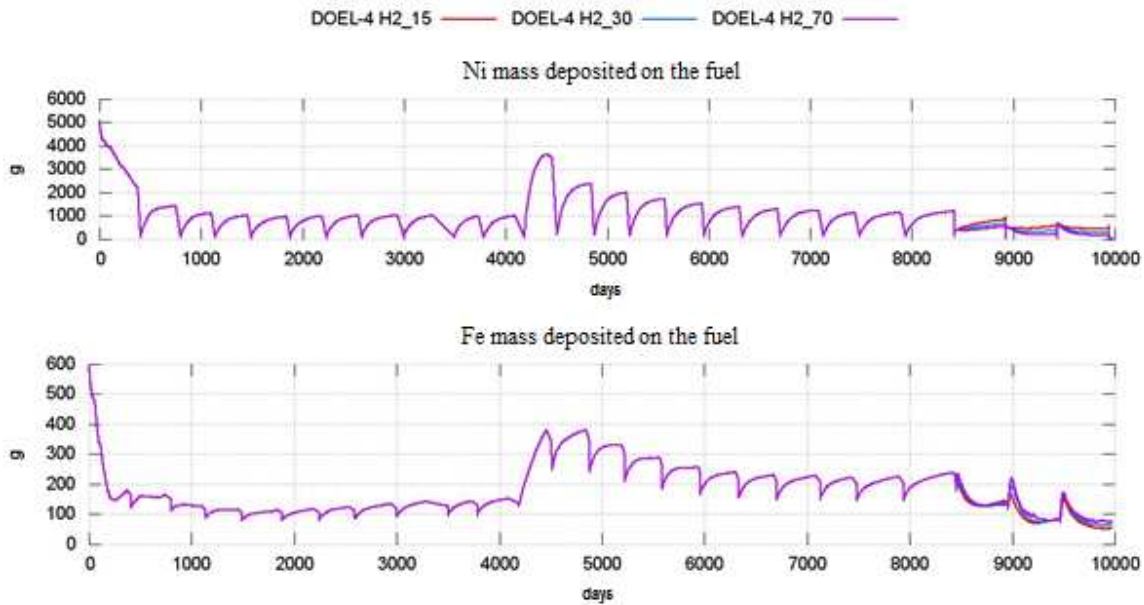


Figure 6: Mass deposited on the fuel

***Evolution of the equilibrium and ionic Ni and Co concentrations inside the reactor vessel and the steam generator tubing***

***Concerning Ni.***

When the DH concentration goes from 15 to 70 mL/kg, equilibrium and ionic Ni concentrations in the core evolve (see Figure 7).

For DH concentrations of 15 and 30 mL/kg, equilibrium Ni concentrations are equal. Ionic Ni concentrations are above equilibrium concentrations which means that Ni tends to precipitate on the fuel and even more for 15 mL/kg (widening of the gap between equilibrium and ionic Ni concentrations) than for 30 mL/kg.

On the contrary, for an increase in the DH value up to 70 mL/kg, equilibrium and ionic Ni concentrations are smaller than those for the two other DH values. Ionic Ni concentrations are lower than the equilibrium concentrations which means that Ni tends to dissolve from the deposit on the fuel into the reactor coolant.



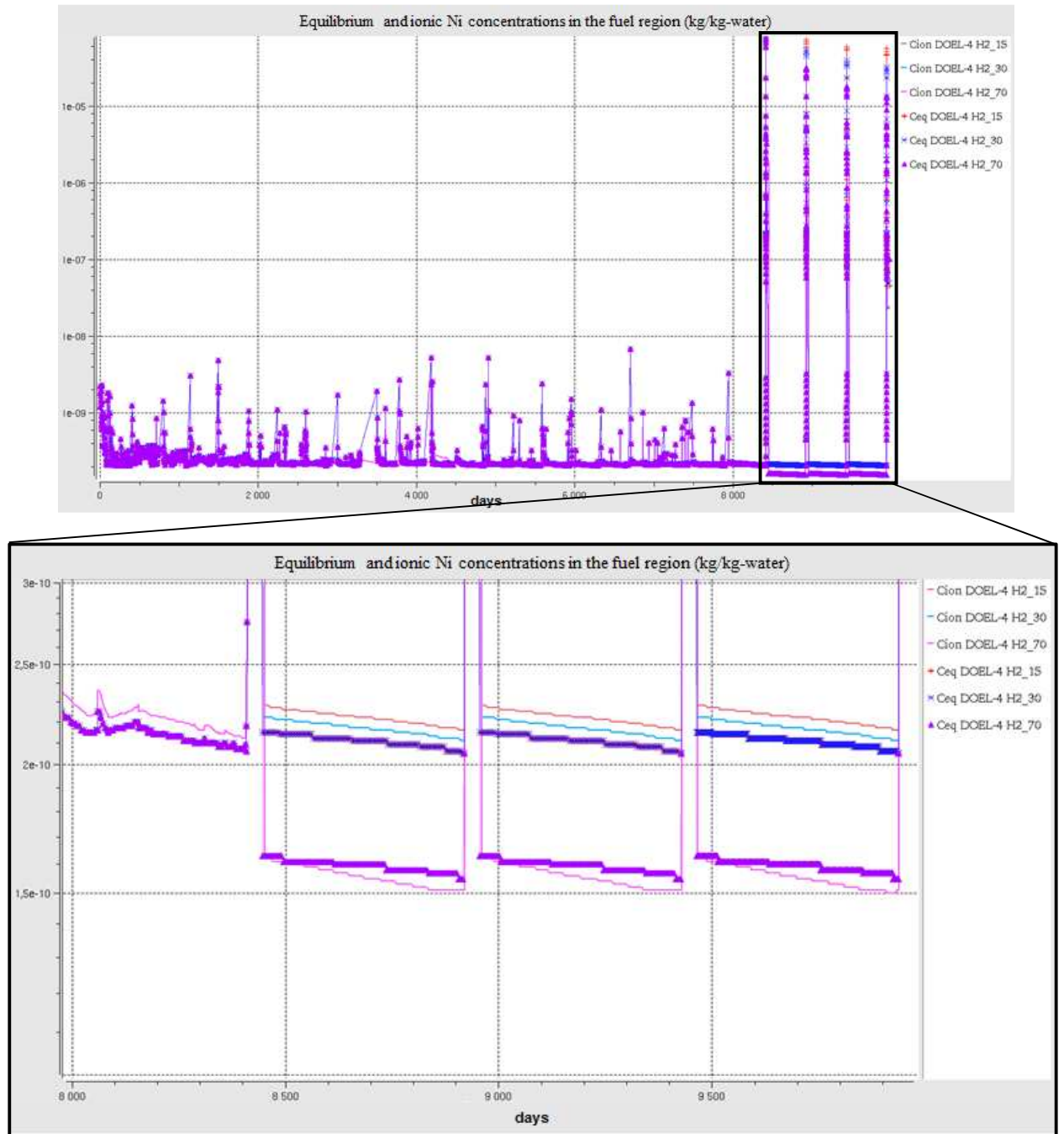


Figure 7: Equilibrium and ionic Ni concentrations in the fuel region

When the DH concentration goes from 15 to 70 mL/kg, equilibrium and ionic Ni concentrations on the cold side of the steam generator tubing evolve (see Figure 8).

Equilibrium and ionic Ni concentrations decrease when the DH increases (For DH concentrations of 15 and 30 mL/kg, ionic Ni concentrations are equal).

For the three DH values, ionic Ni concentrations are much lower than the equilibrium concentrations which means that Ni dissolves in the steam generator tubing in the three cases. An increase in the DH value induces a narrowing of the gap between equilibrium and ionic Ni concentrations and therefore a lower Ni dissolution in the SG tubing for higher DH values.

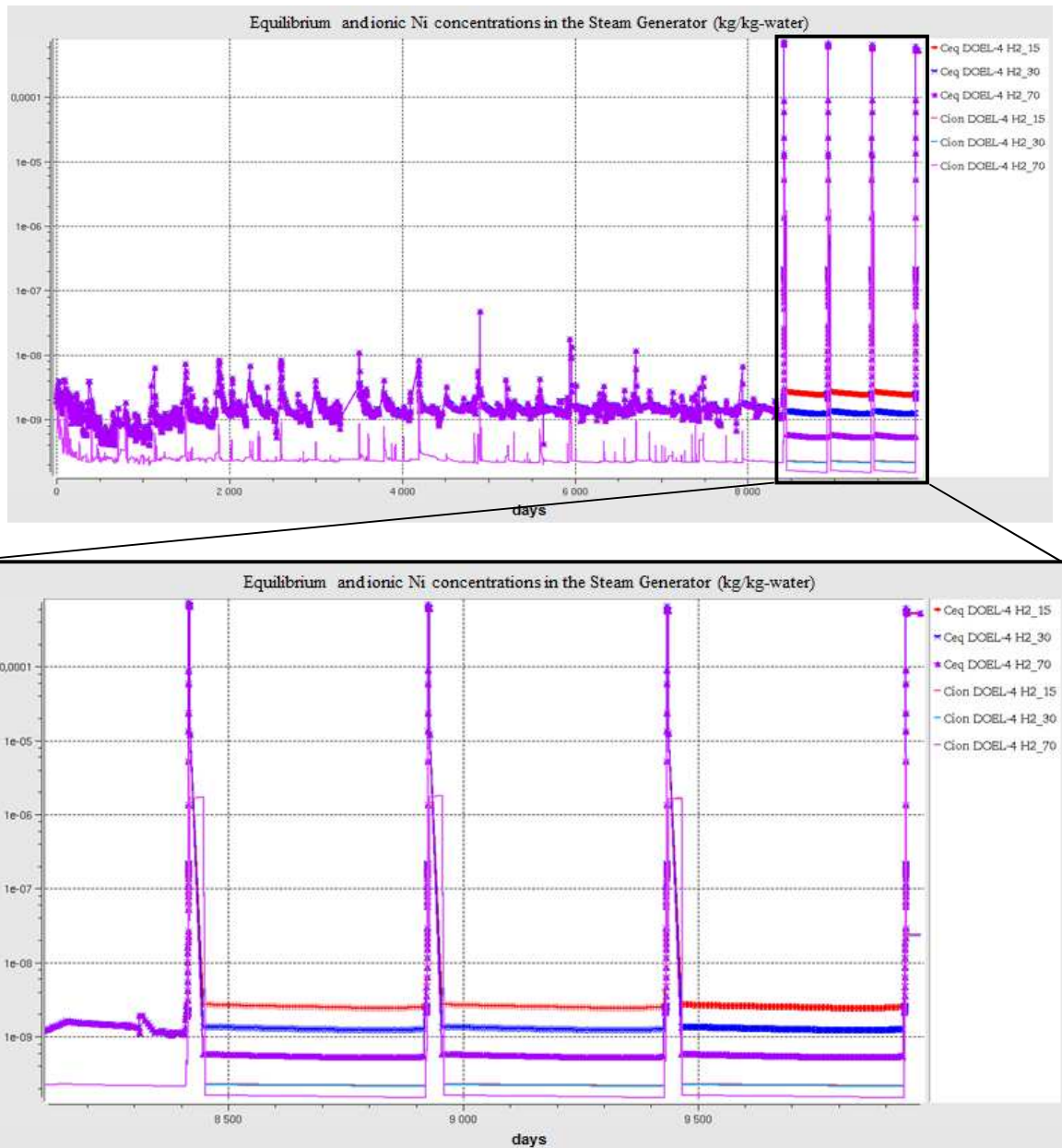


Figure 8: Equilibrium and ionic Ni concentration on the cold side of the steam generator

**Concerning Co.**

Equilibrium and ionic Co concentrations in the fuel regions and the steam generator in the case of a DH value of 70 mL/kg are presented in Figure 9.

Equilibrium Co concentrations are above ionic Co concentrations in the fuel and the steam generator. As a result, Co tends to dissolve and slightly precipitate.

The peaks observed on the last three cycles correspond to the simulated shutdowns.

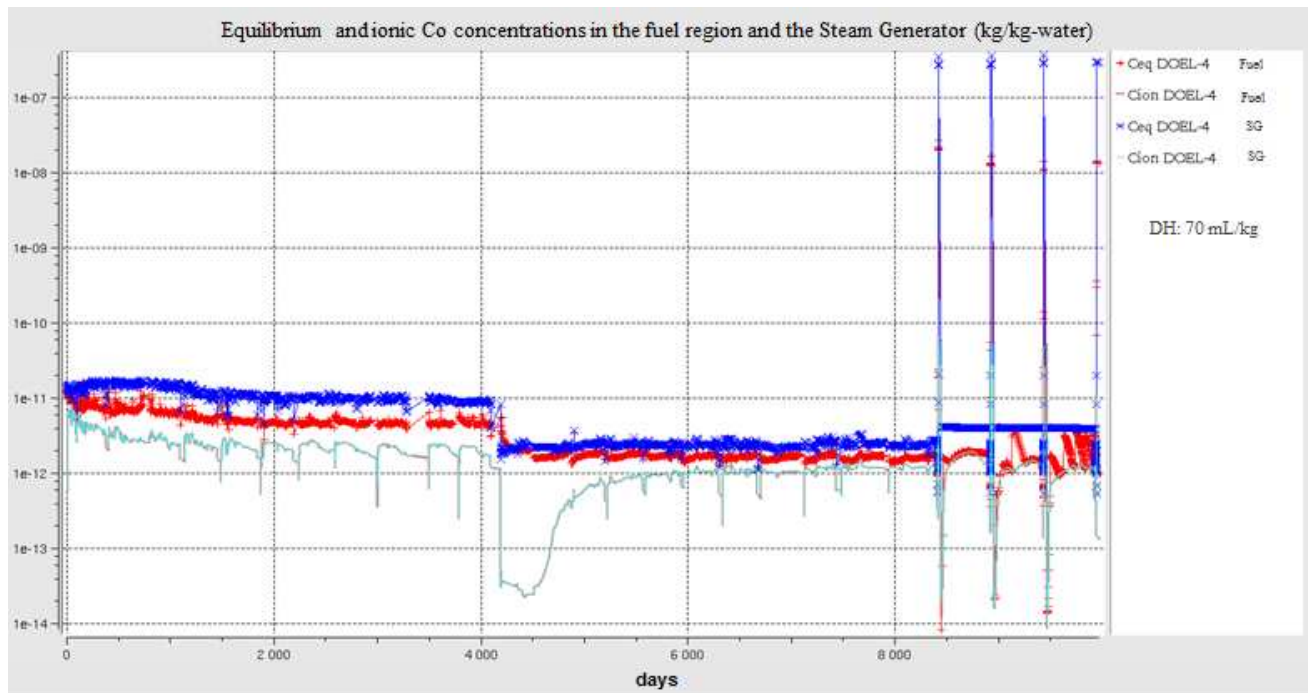


Figure 9: Equilibrium and ionic Co concentrations in the fuel region and the steam generator for a DH value of 70 mL/kg

### ***Ni and Co dissolution versus corrosion release rate in the steam generator***

#### ***Concerning Ni.***

Ni dissolution from the deposit and the release rate from the cold side of the steam generator tubing (for the three DH values 15, 30 and 70 mL/kg) are presented in Figure 10.

Clearly, Ni dissolution from the deposit and corrosion release in the steam generator decrease when the DH concentration increases. The Ni comes mainly from dissolution as the dissolution rate is two orders of magnitude higher than the corrosion release rate.

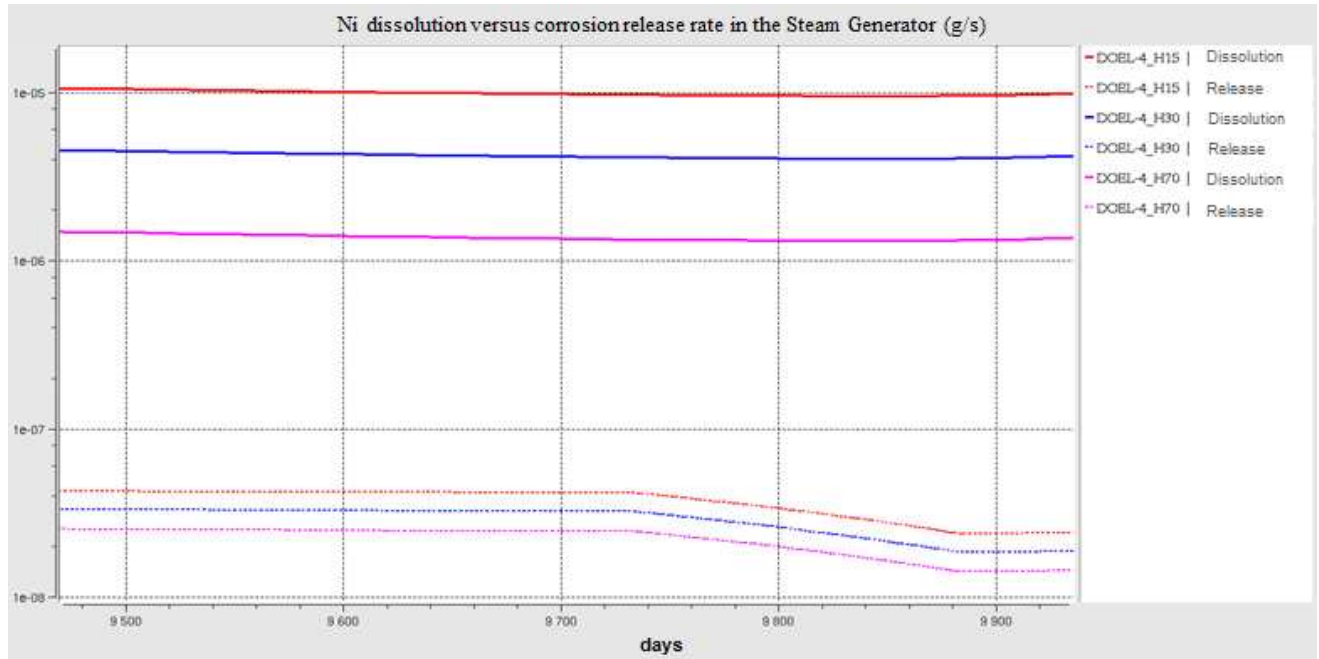


Figure 10: Ni dissolution versus corrosion release rate in the steam generator for the cycle 25

#### ***Concerning Co.***

Co dissolution from the deposit and the corrosion release rate from the cold side of the steam generator (for the three DH values 15, 30 and 70 mL/kg) are presented in Figure 11.

Co dissolution from the steam generator increases when the DH concentration increases. On the contrary, the Co release rate from the steam generator decreases when the concentration increases.

For a DH value of 70 mL/kg, the Co dissolution from the steam generator is higher than for the other DH values while the corrosion release rate is the lowest.

For DH values of 15 and 30 mL/kg, the Co comes mainly from the direct corrosion release from the steam generator.

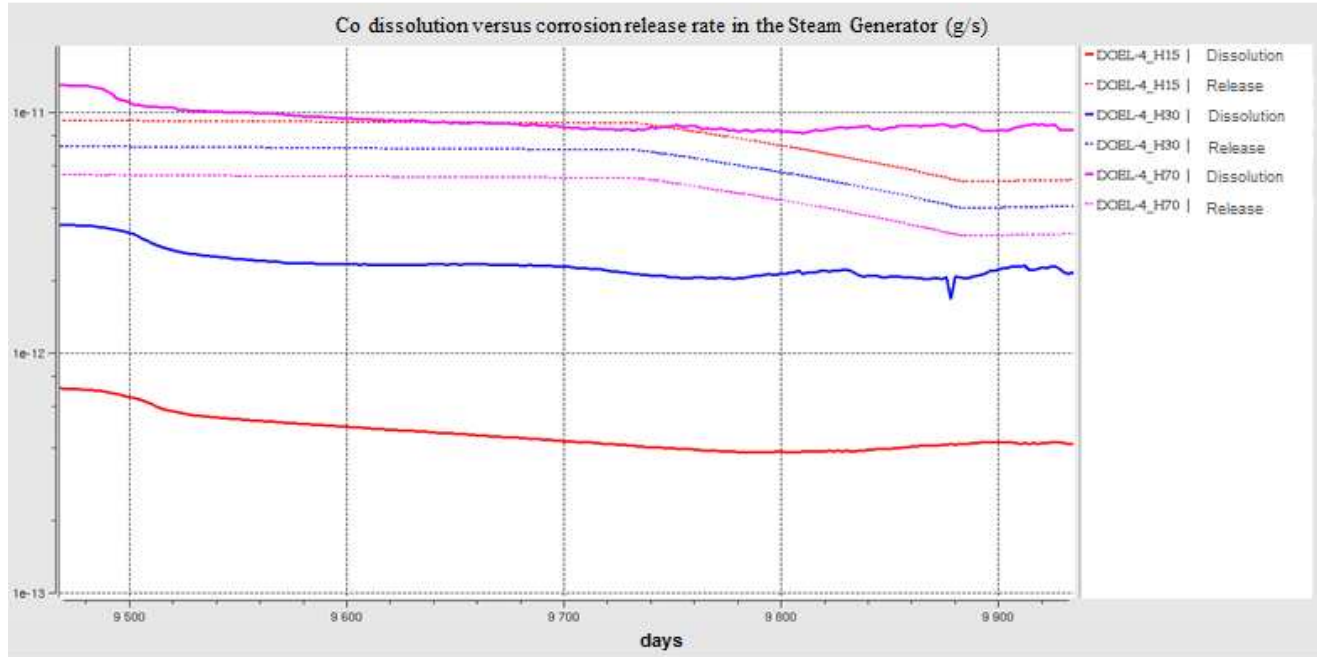


Figure 11: Co dissolution versus corrosion release rate from the steam generator tubing for the cycle 25

For DH concentrations of 15 and 70 mL/kg, the Ni and Co dissolution and corrosion release rate at the end of cycle 25 are compared to those obtained for DH value of 30 mL/kg. The Ni and Co flux ratios at the end of cycle 24 are reported in Table 3.

An increase in the DH concentration up to 70 mL/kg leads to a decrease in the Ni dissolution rate (0.3 factor) and corrosion release rate (0.8 factor) from the steam generator. However, the Co dissolution rate from the SG increases by a factor of 4 while the corrosion release rate decreases by a factor of 0.8.

A decrease in the DH value down to 15 mL/kg leads to an increase in the Ni dissolution rate (2.3 factor) and corrosion release rate (1.3 factor) from the steam generator. However, the Co dissolution rate from the SG decreases by a factor of 0.2 while the corrosion release rate increases by a factor of 1.3.

Table 3: Ni and Co ratios at the end of cycle 25

Ratios / [H <sub>2</sub> ] = 30 mL/kg Cycle 25	Ni rate in the SG		Co rate in the SG	
	Dissolution	Corrosion release	Dissolution	Corrosion release
[H <sub>2</sub> ] = 15 mL/kg	2.3	1.3	0.2	1.3
[H <sub>2</sub> ] = 70 mL/kg	0.3	0.8	4.0	0.8

This table shows that DH concentration significantly affects the Ni rate in the SG, which impacts, due to neutron activation in the fuel regions, the <sup>58</sup>Co contamination of the out of core surfaces.



### DUPLEX experiments

The DUPLEX experimental device [4] is composed of two titanium (TA6V) columns (see Figure 12) crossed by a steady-state and monophasic flow. This device allows testing various DH concentrations. The bottom column, set with a temperature of 270 °C, reproduces the steam generator conditions and contains alloy 690 shavings.

The upper column, set with a temperature of 340 °C, reproduces the fuel cladding and contains pre-oxidized M5 alloy. This upper part is only dedicated to the study of the precipitation of the corrosion products coming from the bottom part.

The elementary composition, especially Ni composition, of different solutions is obtained by mass spectrometry.

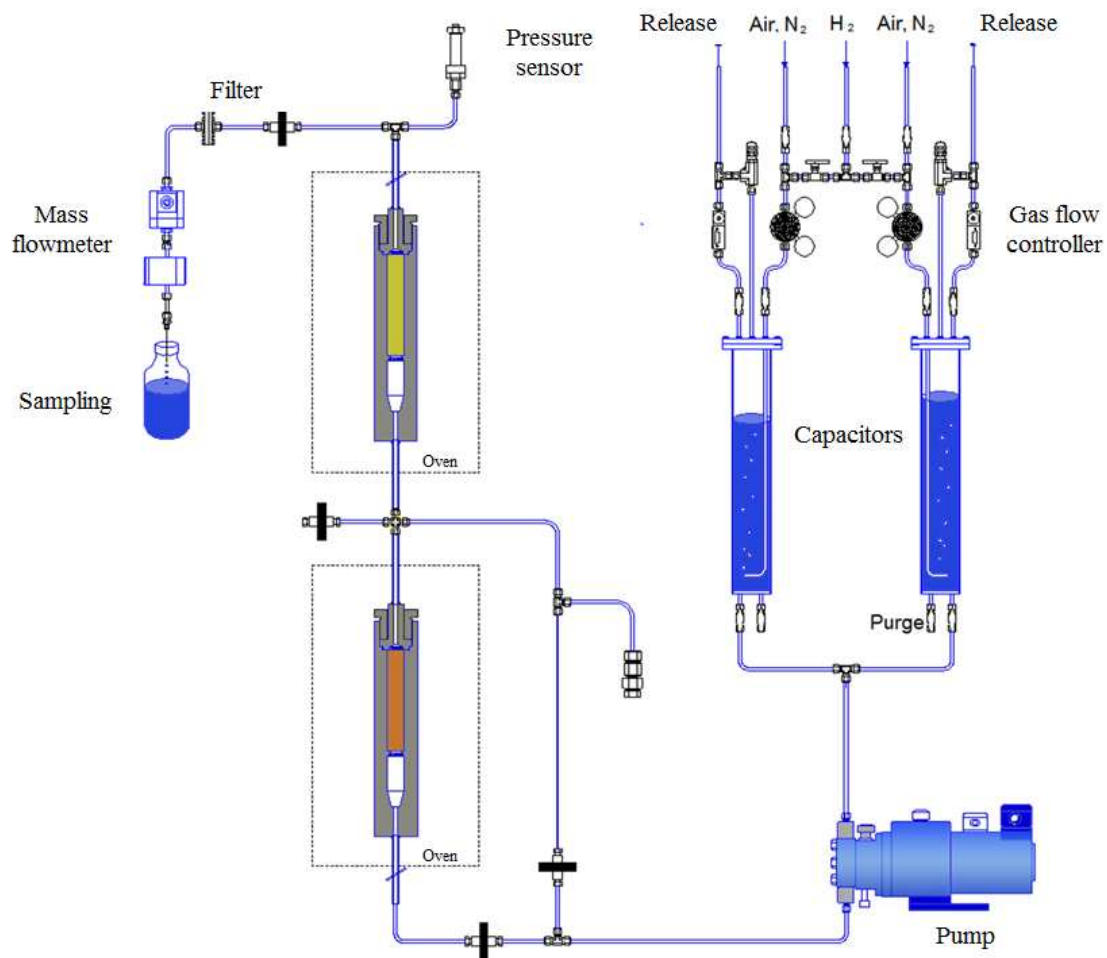


Figure 12: DUPLEX device

The aim of DUPLEX experiments is to study the influence of the DH value (here 10, 28 and 70 cm<sup>3</sup>/kg) on the Ni release coming from alloy 690.

The Ni mass released from alloy 690 is presented for the three DH concentrations in Figure 13. It is clear that the Ni mass decreases when the DH concentration increases. The decrease is even more pronounced when the DH goes from 10 to 28 cm<sup>3</sup>/kg than from 28 to 70 cm<sup>3</sup>/kg.



Those experiments confirmed the OSCAR results obtained in Figure 6.

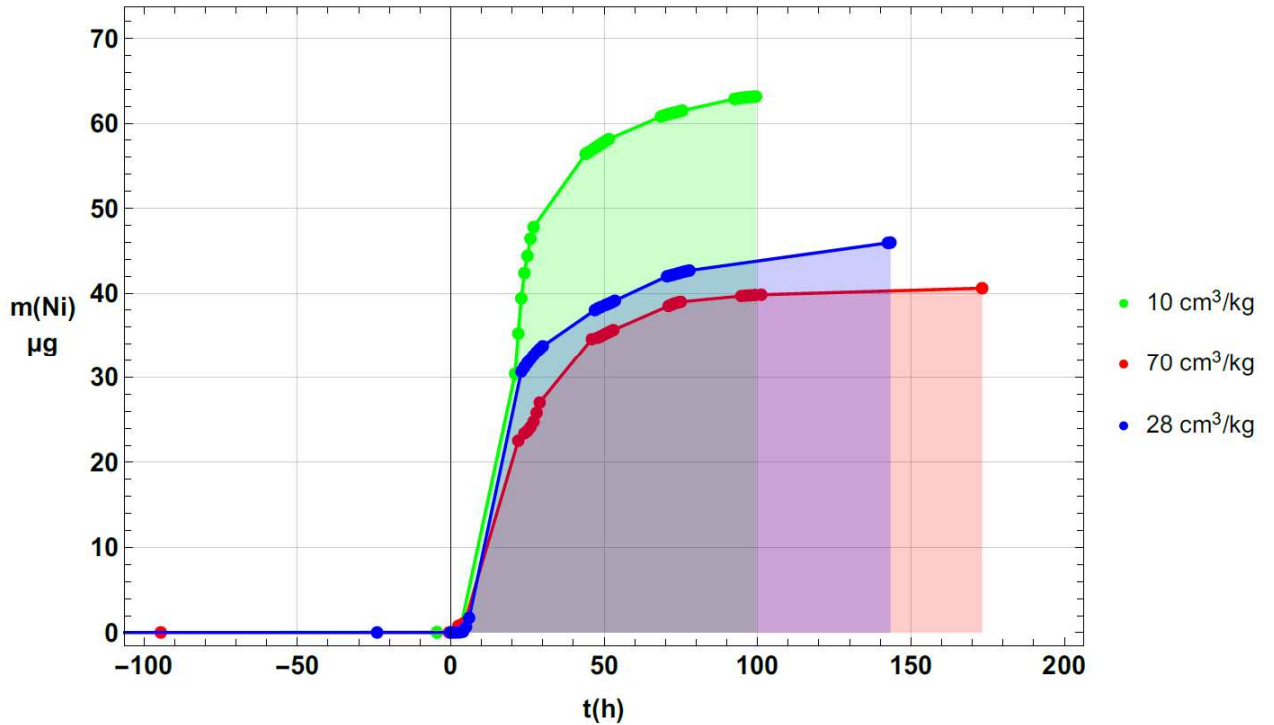


Figure 13: Ni mass released from alloy 690 for various DH values

## Conclusion

For DOEL-4 PWR, calculations have shown that an increase in the DH concentration (from 15 to 70 cc/kg) should result in a decrease in the surface activities of  $^{58}\text{Co}$  and, to a negligible extent, of  $^{60}\text{Co}$ .

Concerning  $^{58}\text{Co}$ , this phenomenon is related to the Ni equilibrium concentration, which decreases (narrowing of the gap between equilibrium and ionic Ni concentrations) in the SG when the DH value increases. This induces a lower Ni dissolution and release from the steam generator for high DH values (70 mL/kg in this study).

This low Ni release from the SG, for high DH values, entails a lower Ni mass that precipitates on the fuel regions leading (by neutron activation of the Ni) to a lower  $^{58}\text{Co}$  contamination.

Concerning  $^{60}\text{Co}$ , equilibrium Co concentrations are above ionic Co concentrations in the fuel and the steam generator regions. As a result, Co tends to dissolve and slightly precipitate.

The Ni mass deposited on the fuel evolves in the same way as the  $^{58}\text{Co}$  surface activity. Indeed, the Ni mass decreases when the DH value increases as observed through OSCAR simulations and DUPLEX experiments

## References

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4. D.You et al., CEA internal report, 2018.