Zinc effect on the primary circuit contamination of a Belgian PWR using the OSCAR V1.3 code

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FROM RESEARCH TO INDUSTRY

Zinc effect on the primary circuit contamination of a Belgian PWR using the OSCAR V1.3 code

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Outline

- Introduction
- Simulation of contamination transfer in presence of Zn using OSCAR
- Analysis of the physicochemical phenomena
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Introduction - OSCAR

OSCAR

Outil de **S**imulation de la **C**ont**A**mination en **R**éacteur

(t**O**ol of **S**imulation of **C**ont**A**mination in **R**eactor)

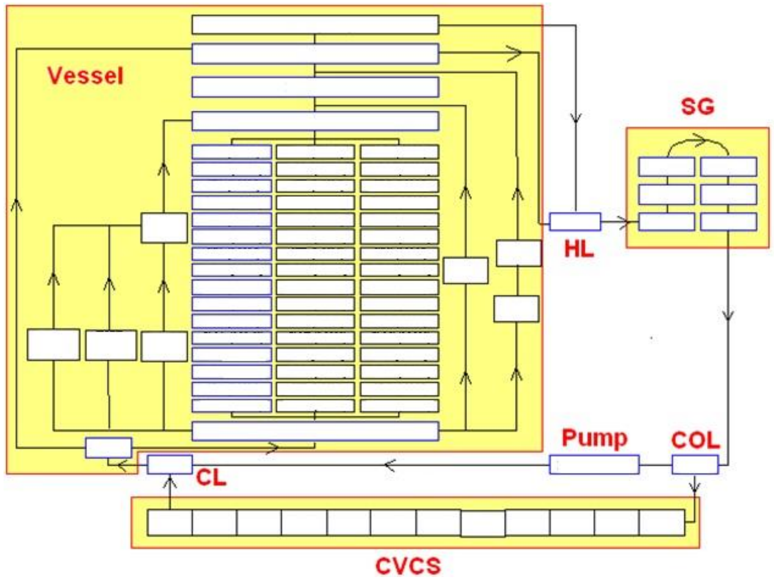
- ✓ Simulation of contamination transfer in nuclear reactor systems during power operation and during cold shutdown
- ✓ Calculation of masses/activities of corrosion products, activated corrosion products, fission products and actinides in solid, liquid and gaseous phases of nuclear circuits as function of time (normal operation over several decades and transients over several minutes/hours)
- ✓ Developed since 70's (formerly PACTOLE and PROFIP codes) in a collaboration CEA – EDF – AREVA

Introduction – OSCAR V1.3 Corrosion Products

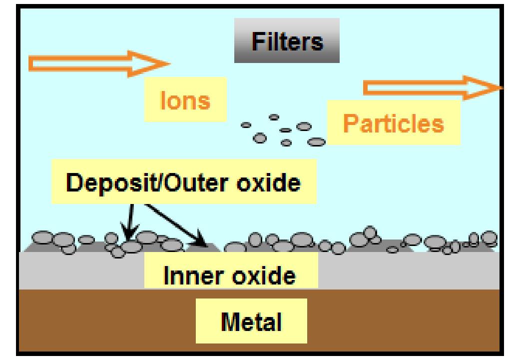
❑ Circuits discretized in control volumes

according to:

- material
- geometry
- thermal-hydraulics
- neutronics
- operation



❑ Up to 6 media in each control volume



❑ Elements/Radioisotopes:

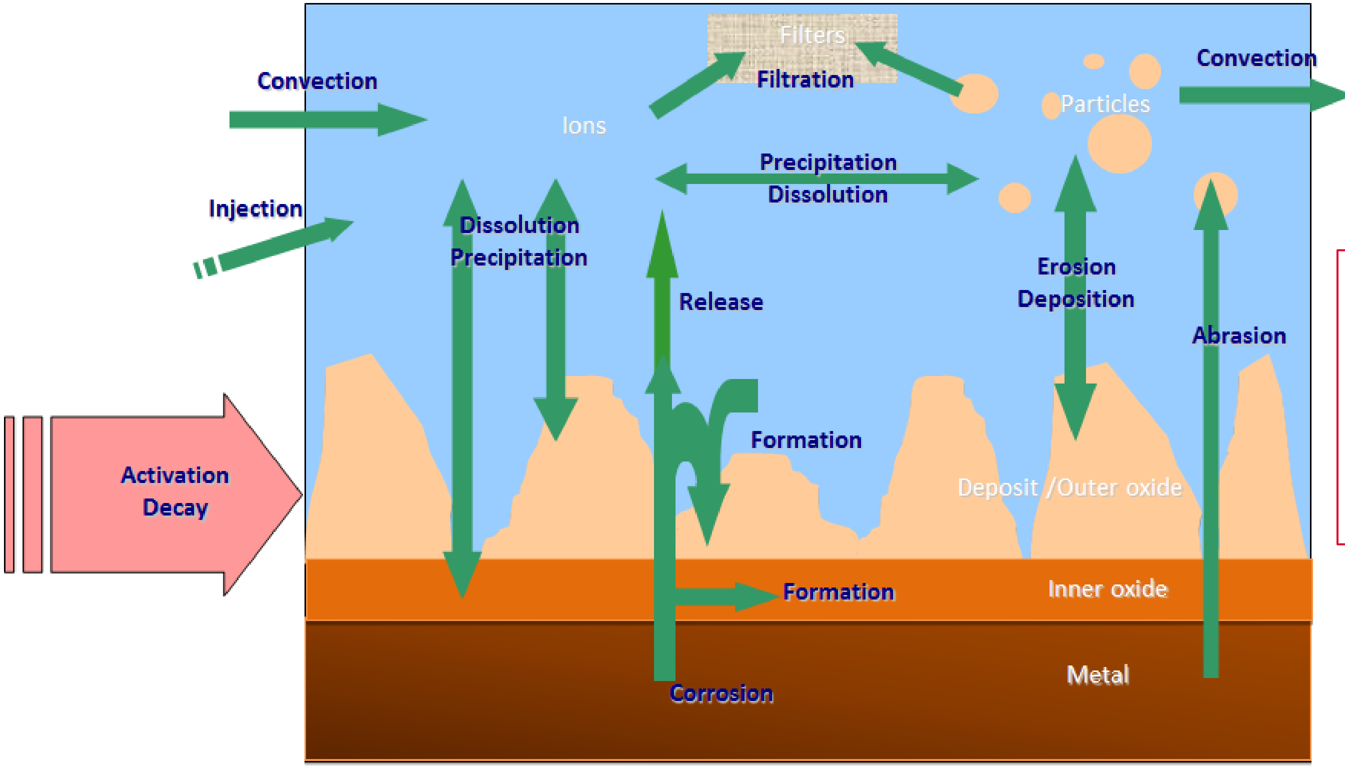
- Up to 8 elements: Ni, Co, Fe, Mn, Cr, Zr, Ag and Zn
- Up to 15 radioisotopes: ^{58}Co,^{60}Co, ^{54}Mn, ^{51}Cr, ^{65}Zn…

❑ Instantionnary mass balance equation for each isotope in each medium of each region:

$$\frac{\partial m_i}{\partial t} = \sum_{Source} J_m - \sum_{Sink} J_m$$

- m_i : mass of isotope i in a medium
- J_m : mass flux between 2 media or 2 isotopes or 2 regions

Introduction – OSCAR CPs – Transfer mechanisms



Region k

- Possibility to inject Zn
- Zn is subject to transfer mechanisms, in particular to dissolution/precipitation mechanism

➢ Dissolution/Precipitation rate [kg.m^{-2}.s^{-1}]:

$$J^{elt} = \frac{1}{R} \cdot (C^{elt}_{equil} - C^{elt})$$

- R : mass transfer resistance between oxide and bulk
- C^{elt}_{equil} : equilibrium concentration of element *elt*
- C^{elt}: bulk concentration of element *elt*

C^{elt}_{equil} and composition of ideal solid solution (mixed oxides and pure solid phases in excess) calculated by PHREEQCEA and its thermodynamic database (OSCAR chemistry module)

Introduction – Objective

- Zinc injected in PWR primary system in order to reduce ^{60}Co contamination
- Great Zn affinity for chromites (inner oxide)
- Experience feedback of Zn injection:
  - Slight variations of ^{60}Co contamination of old oxidized surfaces (**EMECC** measurements in 8 different PWRs in France and abroad)
  - Higher ^{60}Co volume activity during cycle (not always)
  - Higher ^{60}Co activity deposited on fuel rods

- **Objective of this study:**
  - Compare OSCAR simulation with Zn to experience feedback
  - Progress in understanding phenomena involved in contamination in presence of Zn

EMECC campaigns: Measurements by gamma spectrometry of activities deposited inside components (pipes, steam generator tubing, heat exchangers…):

- ~380 campaigns in 70 different PWRs since 1971

EMECC device
Measurement of a PWR hot leg

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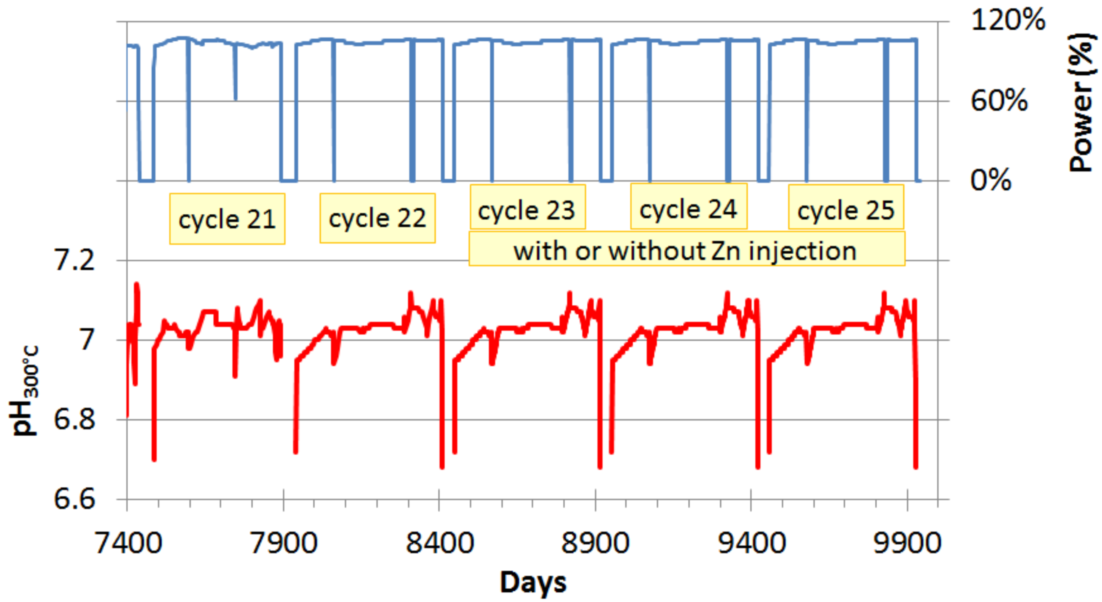
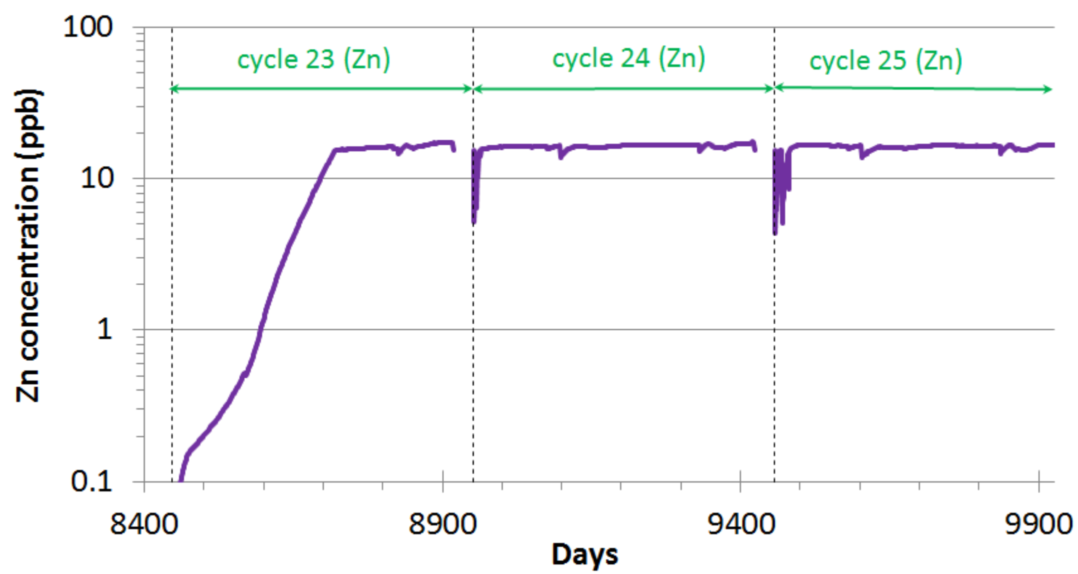
Simulation – Zinc injection

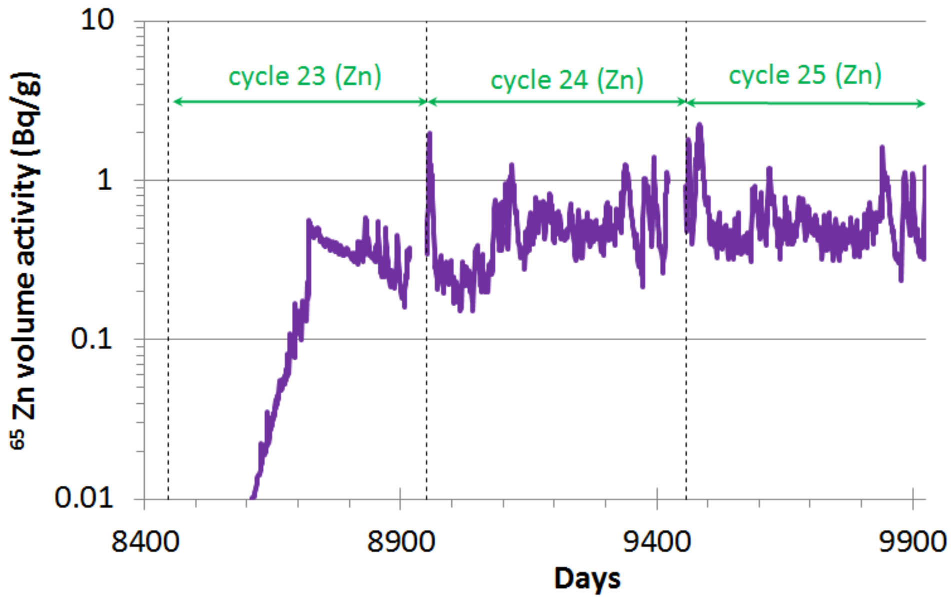
- OSCAR simulations:
  - 3-loop Belgian PWR
  - 22 cycles without Zn injection
  - Cycles 23 to 25 :
    - Same operating parameters as cycle 22 with or without Zn injection
    - Zn injection:
      - $[Zn]_{target}$ = 15 ppb
      - Depleted in ^{64}Zn

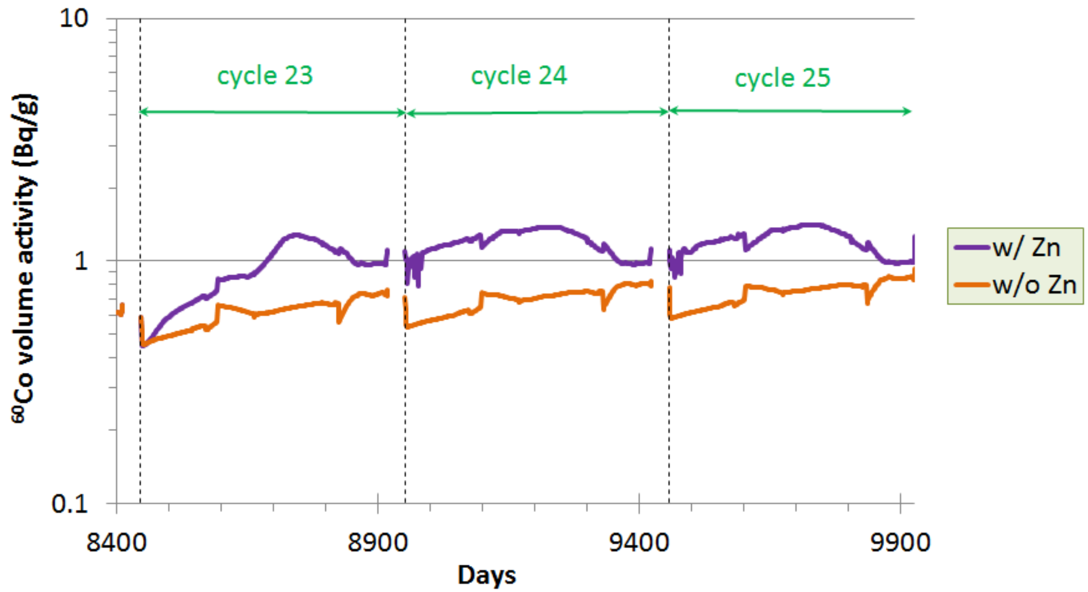
Cycle	Zn injection rate
23	65 g/day (9 months) 13 g/day (11 months)
24	12 g/day
25	11 g/day





Simulation – Volume activities

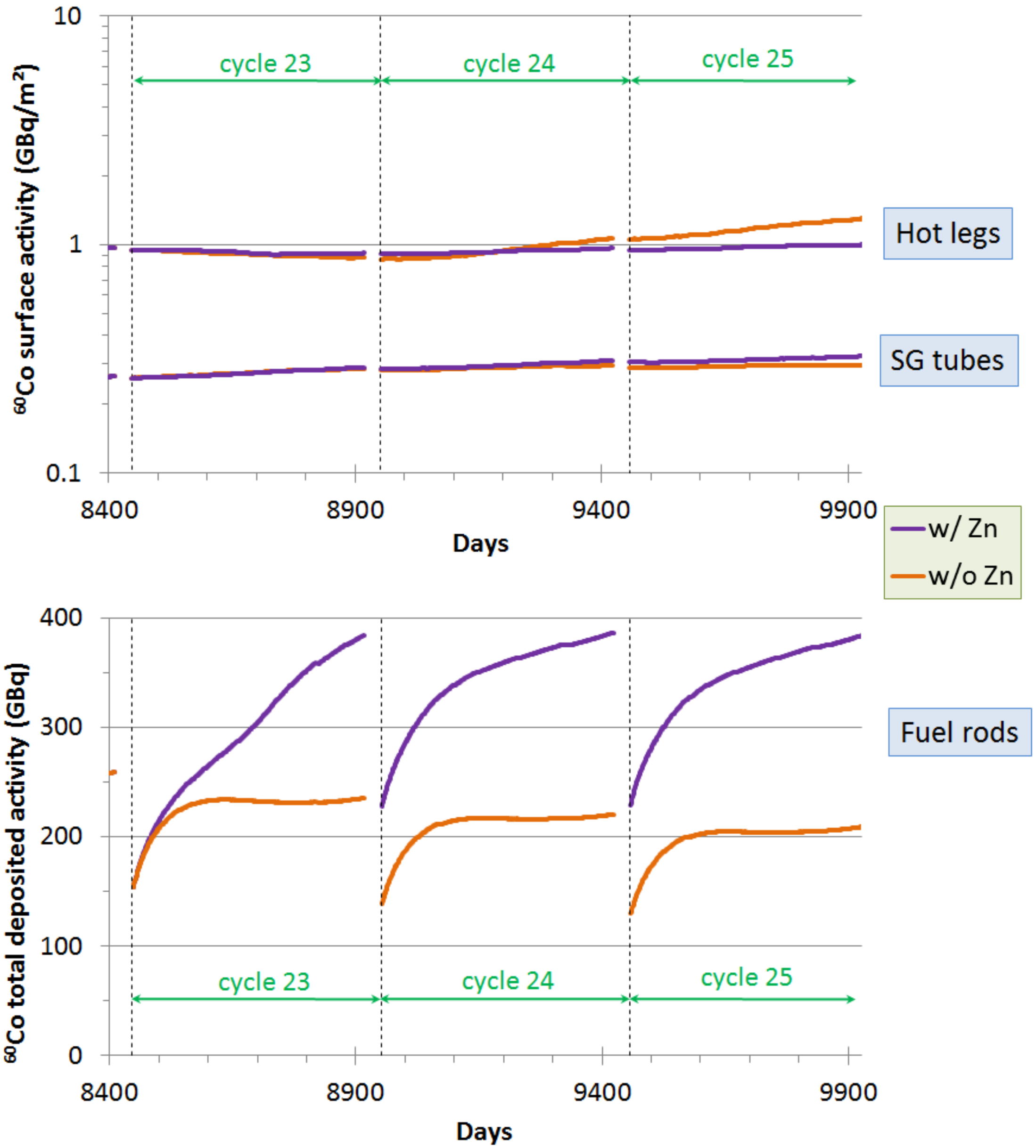




- Impact of Zn injection:
  - ^{65}Zn volume activity: 0.2-2 Bq/g
  - Increase in ^{60}Co volume activity by a factor ≈2

Consistent with experience feedback

Simulation – ^{60}Co deposited activities



- Impact of Zn injection: depends on region and cycle
  - Out-of-flux surfaces:
    - At the end of first cycle: no significant difference
    - At the end of third cycle: increase in ^{60}Co surface activity (+10% to +30%) except on hot legs (-20%)
  - Fuel rods:
    - Increase in ^{60}Co total deposited activity (+60% to +80%)

Consistent with experience feedback

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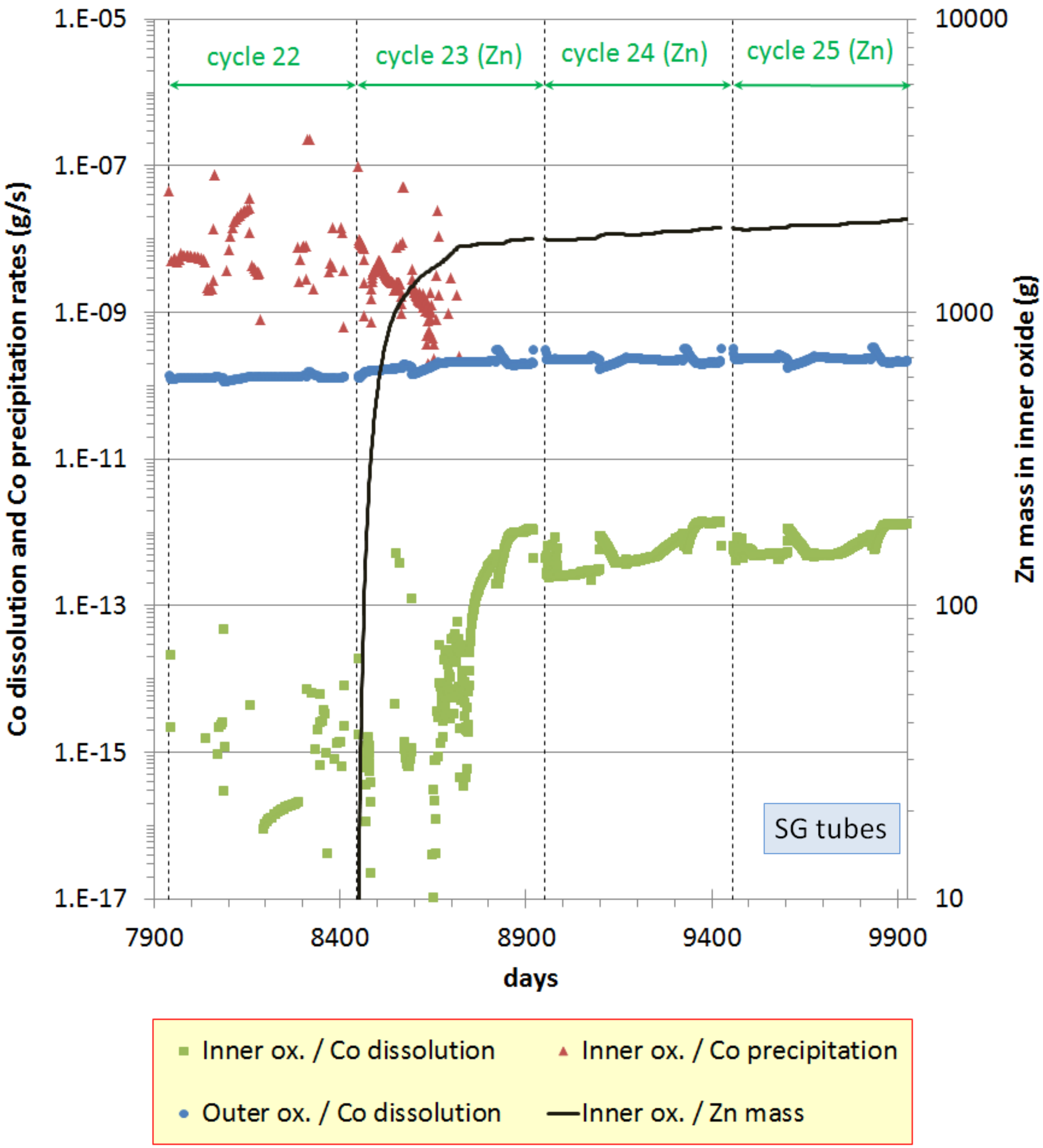
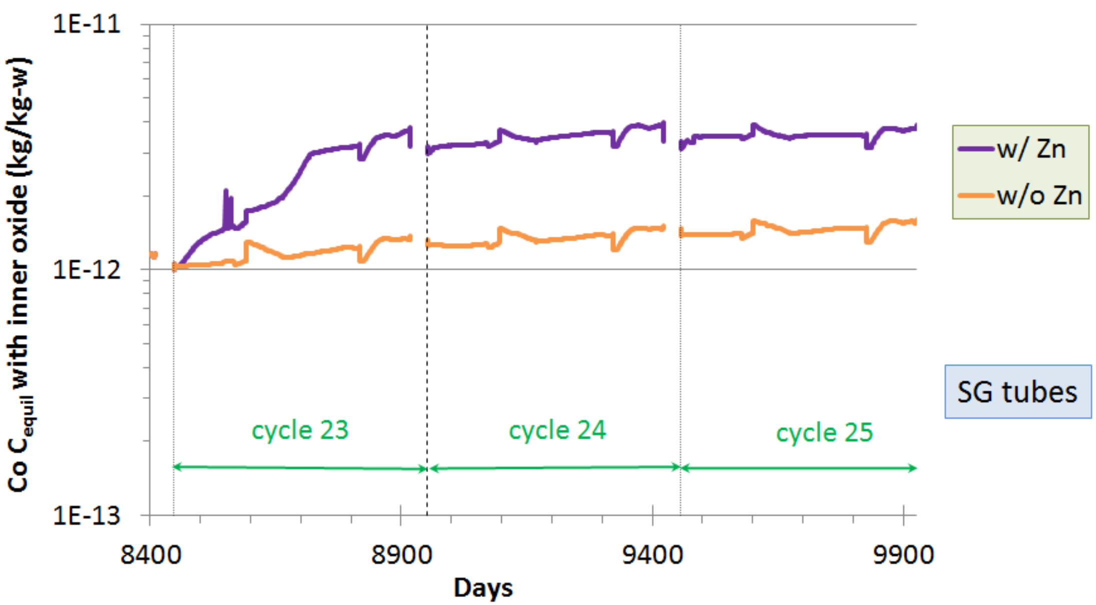
Simulation – Zinc injection - Explanation



- Strong precipitation of Zn on inner oxide until saturation
  - → Consistent with great Zn affinity with chromites
- When inner oxide saturated by Zn:
  - No Co precipitation anymore on inner oxide
  - Higher Co dissolution (10-100 times higher)
- Practically no Zn impact on Co dissolution from outer oxide

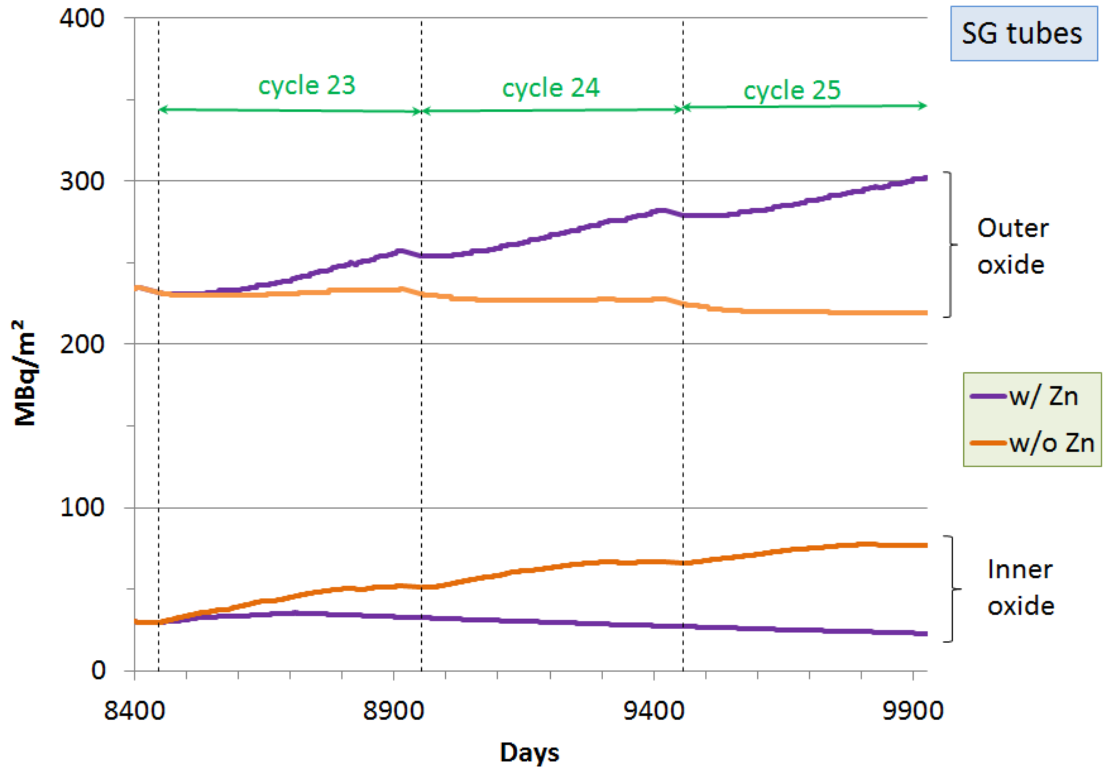
Simulation – Zinc injection - Explanation



- Strong precipitation of Zn on inner oxide until saturation
  → Consistent with great Zn affinity with chromites
- When inner oxide saturated by Zn:
  - No Co precipitation anymore on inner oxide
  - Higher Co dissolution (10-100 times higher)
- Practically no Zn impact on Co dissolution from outer oxide
- Change in stability of the Cr-rich solid solution (inner oxide) due to Zn
  - Increase in C^{Co}_{equil} calculated by OSCAR chemistry module (PHREEQCEA)

Simulation – Zinc injection - Explanation

- No more Co precipitation on inner oxide and Co dissolution from inner oxide
  - **Decrease in ^{60}Co contamination of inner oxide (=decay rate)**
  - Surplus of Co deposited on fuel rods
  - Increase in ^{60}Co activity on fuel rods
- Erosion of deposit on fuel rods
  - Increase in ^{60}Co particle volume activity (also due to ^{60}Co dissolution from inner and outer oxide)
- Deposition of particles on outer oxide of out-of-flux components
  - **Increase in ^{60}Co contamination of outer oxide**



Antagonist effects: explanation of slight impact observed by EMECC campaigns

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Conclusion

- **OSCAR**: useful to progress in understanding contamination transfer in nuclear circuits
- According to OSCAR V1.3, slight impact (beneficial/detrimental) on ^{60}Co surface activity of old oxidized surfaces due to antagonist effects:
  - Decrease in ^{60}Co contamination of inner oxide
  - Increase in ^{60}Co contamination of outer oxide
  - Explanation in accordance with assessment of **EMECC** campaigns in PWRs with Zn injection
- Note:
  - No specific mechanism added to OSCAR to reproduce Zn effet
  - Only Zn effect on chemical equilibrium taken into account
    No Zn effect on corrosion in OSCAR V1.3
    → Taken into account in OSCAR V1.4 released in 2017

Thank you for your attention

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