Overview of French R&D studies for the development of radwaste packages limiting tritium outgassing

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■ Context

Context and origin of the need

- **The radwaste management approach proposed by a waste producer relies scenarios taking into account:**
  - The nature of the waste (radiological content and physico-chemical characteristics)
  - The estimation of waste amount production and the associated chronicle
  - The existing waste management facilities and disposals able to handle the expected waste

- **In case of fusion facilities using Deuterium (D) and Tritium (T) as fuel, a large part of the waste is expected to be tritiated with very different level of T content**

- **Among them, the relatively highly tritiated waste could require a specific management approach because**
  - Tritium behaviour in materials
  - Such waste were not produced in such large proportions in the current nuclear industry
  - And thus the current acceptance criteria regarding tritium of the facilities foreseen to manage radwaste could be exceeded or the tools to be used possibly not exist.

Context and origin of the need

- **The need of a specific management approach results largely from the relatively high tritium content in the waste and the ability of tritium to permeate through materials**

- **Considering these 2 aspects, a specific management approach for the highly tritiated waste could rely on different options:**
  - The minimization of tritium contamination at source
  - The reduction of tritium content in the produced waste by specific treatment abling the Tritium recovery
  - The limitation of Tritium degassing out of the waste package (T outgassing)
  - The limitation of the Tritium transfer in the foreseen waste management facilities (mainly treatment facilities and disposal facilities)

- **This lecture will focus on the workplan developed by CEA to tackle the issue related to tritium outgassing from waste packages**

1. Issues related to tritiated waste management

Characteristics of tritiated radwaste possibly produced by fusion facility

- **In order to define the requirements and the level of performance of the solutions to be developed to limit tritium outgassing from the waste package, it is mandatory to define a reference case with the typology of waste that can be produced by a fusion facility**

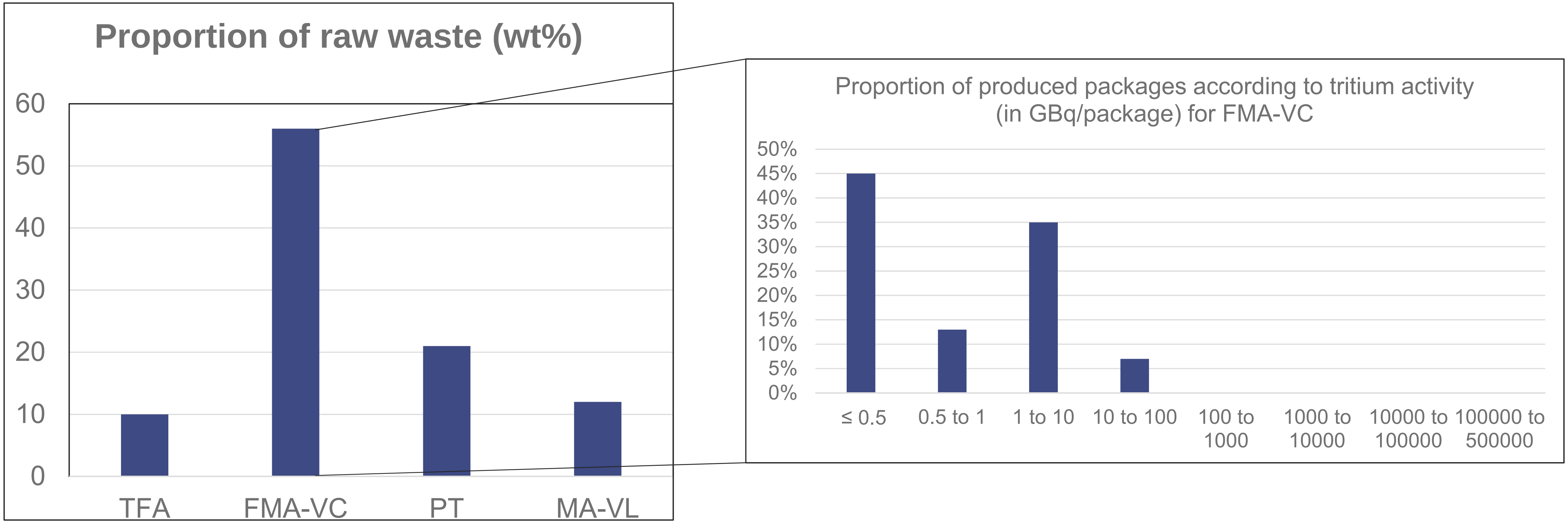
- **For that, ITER waste inventories and characteristics estimations were used even if some studies are still ongoing to refine them**

- **For a fusion facility like ITER, the produced waste are from the following categories according to French nuclear waste package classification:**
  - TFA waste (French abbreviation meaning very low-level),
  - FMA-VC waste (French abbreviation meaning low and intermediate-level short-lived),
  - MA-VL waste (French abbreviation meaning intermediate-level long-lived).

  And another category is specifically mentioned: the purely tritiated (PT) waste containing only Tritium as radionuclide

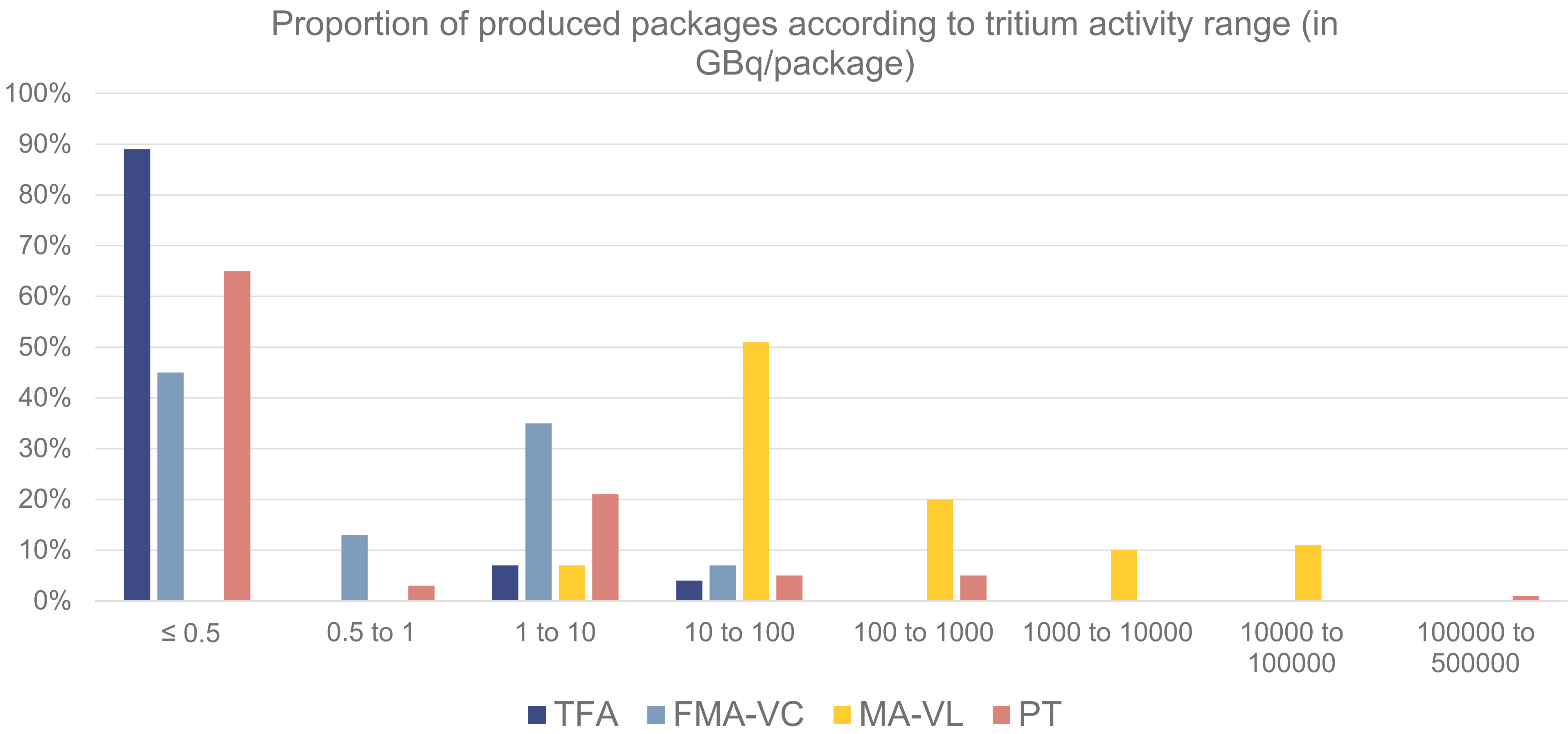
Characteristics of tritiated radwaste possibly produced by fusion facility

- **Example of ITER radwaste typology (based on the current estimations)**



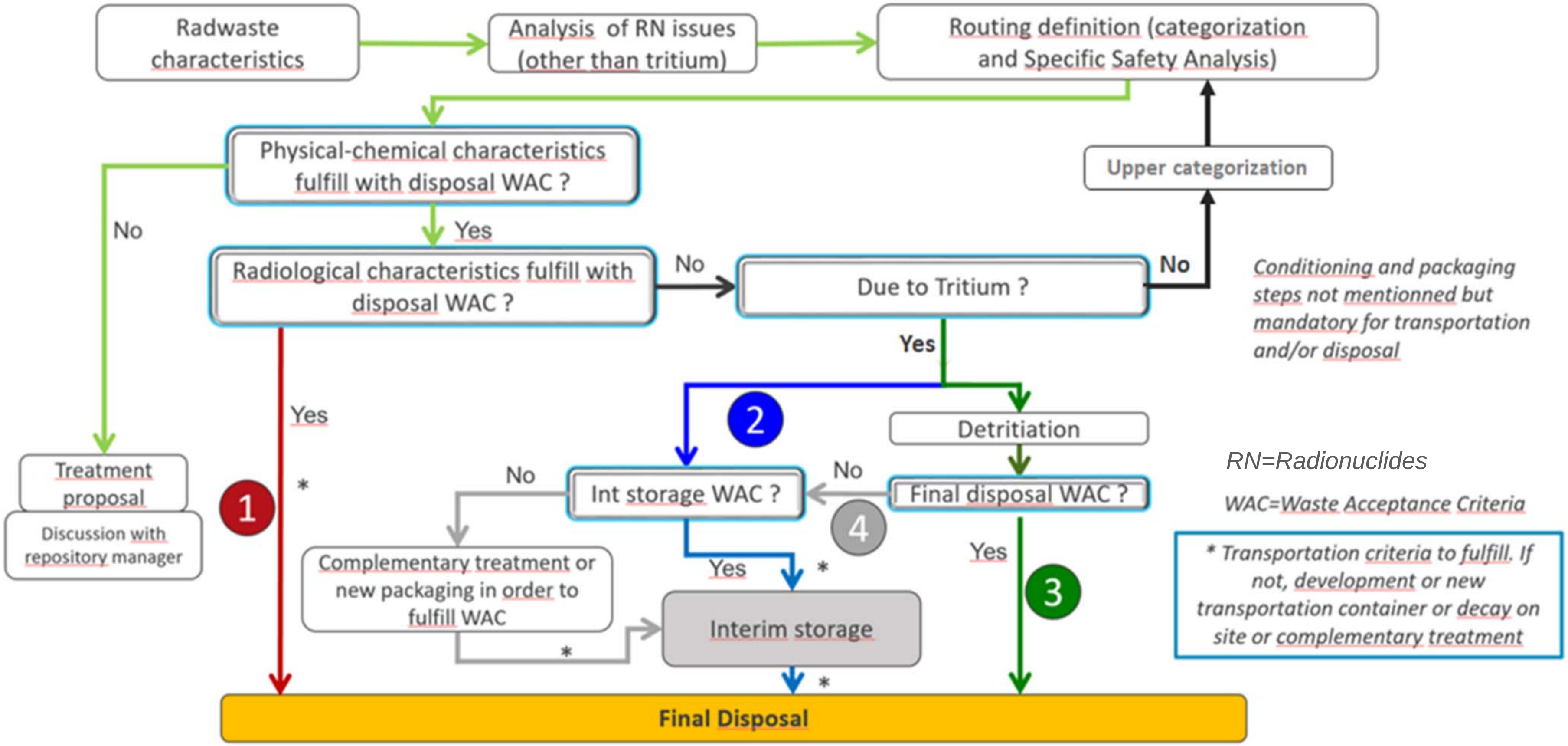
Characteristics of tritiated radwaste possibly produced by fusion facility

- **Example of ITER radwaste typology (based on the current estimations)**



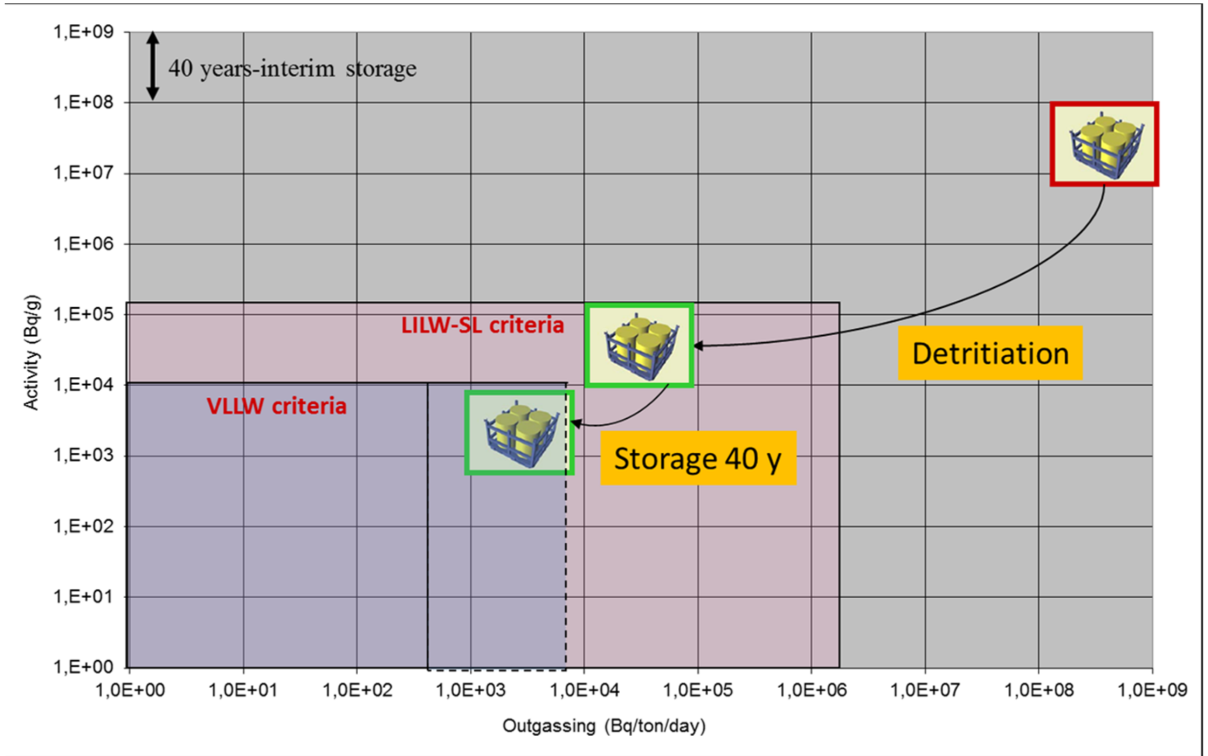
Main options for a tritiated waste management strategy

- Current scheme of decision for the strategy definition



Main options for a tritiated waste management strategy

- **Without any treatment, some waste will have to be stored for tritium decay during decades prior being compliant with final repositories WAC**



- **But for the highest tritiated waste, 40 years or 50 years of interim storage could not be enough to fulfill the waste acceptance criteria of the existing final disposal**

- **Thus, packages limiting T releases could represent an opportunity to reach tritiated waste acceptance criteria**

3. Confining waste package development

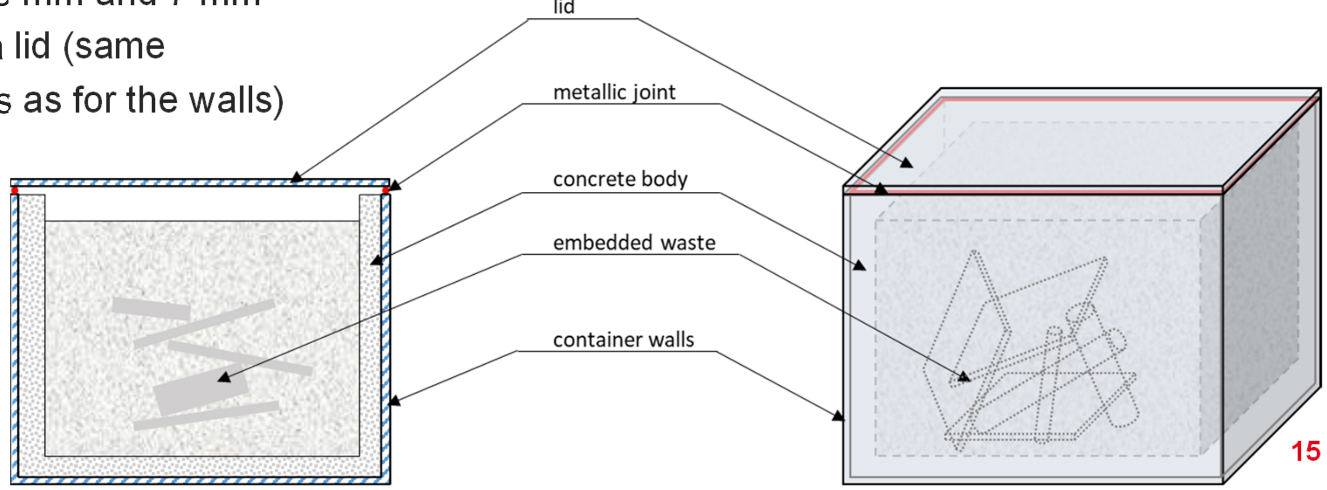
Development of package limiting tritium outgassing

- **The steps of development of packages limiting tritium outgassing relies on :**
  - The evaluation of the capabilities of metallic liner in the container
  - The selection of an appropriate seal
  - The selection of getter for tritium trapping
  - The selection of a blocking matrix compatible with the waste and the getter
  - The demonstration of the confining performance of the getter in representative conditions
  - The extrapolation of the performance over decades → development of a representative modelling

Metallic liner and seal impact

Material and characteristics of the container: metallic liner impact on T outgassing

- **Objective: tritium outgassing evaluation with different metallic liner in the waste package**
- **A possible option to enhance the performance of the package in order to limit T outgassing consists in the use of a metallic liner in the waste package (concrete presents poor confinement performance towards tritium even though an effort is made for the choice of the cement matrix)**
- **In the purpose of preliminary evaluations of container performance, we defined main assumptions concerning:**
  - Container characteristics:
    - parallelepipedic container with stainless steel walls (304L)
    - Different wall thicknesses : 5 mm and 7 mm
    - The container is closed by a lid (same material and same thickness as for the walls) sealed by a metallic joint (under investigation) or welding (or



Material and characteristics of the container: metallic liner impact on T outgassing

- **In view of preliminary evaluations of container performance, main input data and assumptions are :**

  - Specific waste with relatively high tritium content:
    - 2.81 x 10^7 Bq/g corresponding to waste from the divertor after an interim storage of 50 years and without any detritiation treatment.
    - 1 x 10^5 Bq/g corresponding to the average activity of tritiated waste from ITER.

  - The T outgassing rate from the raw material leading to a partial pressure in the void spaces of the container:
    - The metallic waste outgassing rate (20% of the inventory per year).
    - The concrete matrix of embedded waste and reinforcing the walls of the container is not considered as a permeation barrier.
    - The void fraction (fraction of the volume occupied by the matrix) in the container is 25 %.
    - The temperature is considered to be homogeneous and is the same in all the walls.

  - Waste package storage conditions: calculations were performed at 2 different temperatures, 20°C and 30°C corresponding to possible temperature of the final disposal

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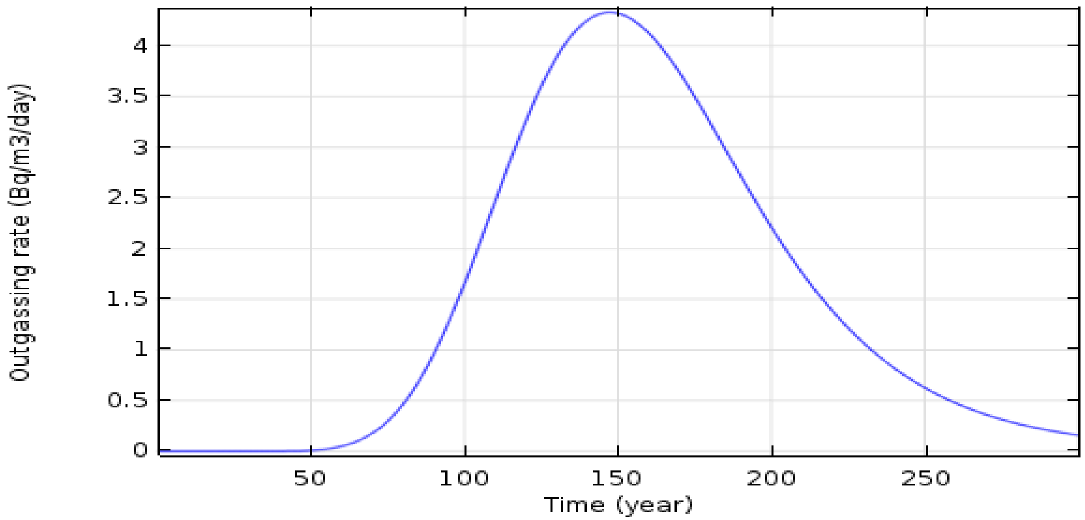
Material and characteristics of the container: metallic liner impact on T outgassing

- **Preliminary results:**
  - The maximum outgassing rate around 4 Bq/m^3 of container/day is observed after 146 years.



Evolution of the tritium outgassing rate.

- For any studied cases, the tritium outgassing rates calculated from the tritium diffusion through the container walls are very low and far below the limit fixed in the WAC of the French Final disposal devoted to FMA-VC
- Over the whole range of parameters used in this study, the maximum outgassing rate is reached after a time going from 146 years to 288 years.

Material and characteristics of the container: seal impact on T outgassing

Assessment of the leakage through the metallic seal

- For such low temperatures and with wall thicknesses of at least 5 mm, tritium permeation through the walls of the container might not be of major concern regarding the overall tritium releases
- A contribution to these tritium releases could also be the tritium leaks through the container seal
- Existing closure systems used for tritiated waste containers have leak rates between 10^{-9} and 10^{-7} Pa.m^3.s^{-1} leads to the following range of tritium leak rates from packages:

Leak rate [Pa.m^3.s^{-1}]	10^{-7}	10^{-9}
T leak rate [Bq/m^3/day]	1.90×10^{6}	1.90×10^{4}
T leak rate [Bq/t/day]	6.24×10^{5}	6.24×10^{3}

In red : above the WAC
In green : below the WAC

Assessment of the tritium releases for different leak rates for the most pessimistic case regarding T outgassing (5 mm / 28 MBq.g^{-1} / 30°C).

- Highly efficient metallic joint with a leak rate smaller than 10^{-7} Pa.m^3.s^{-1} is required to limit the tritium release from the container.
- Moreover, the selected joint will have to guarantee its performance over a long period of time.

- Another limitation could come from the presence of cracks in the welded parts of the metallic wall
  → **a complementary solution for tritium outgassing rate limitation could be needed**
    → **trapping in the cement matrix**

Cement matrix and Tritium getter impact

Development of specific packages - $HT/T_2/H_2$ getter selection

- **Trapping systems that are able to limit hydrogen or tritium release**
  - Organic
  - **Inorganic (oxides)** → γMnO_2 /Ag_2O getter
    (≈87 wt.%) (≈ 13 wt%)
  - Hybrid

- Better resistance to irradiation than organic getters
- Stable in air or under water unlike certain metal hybrids
- Reasonable cost due to the replacement of noble metals (Pd, Pt…) by Ag_2O as a promoter
- Irreversible trapping of dihydrogen

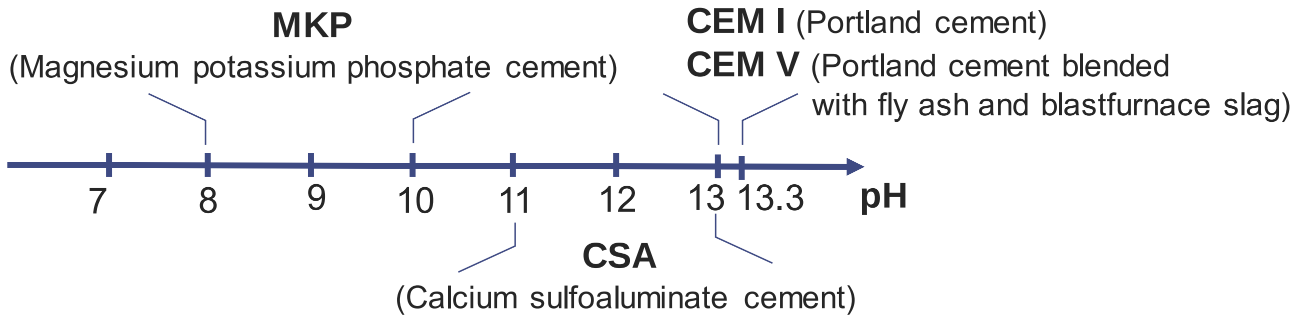
(Kosawa, 1981; Chabre, 1995; Maruejouls, 2003)

- **Identification of the possible trapping mechanism**

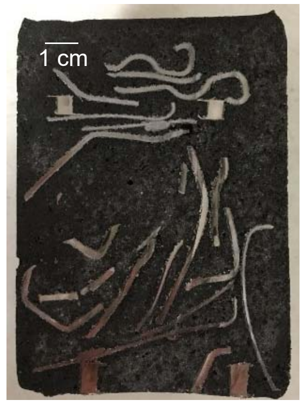
Development of specific packages- Formulation of cement-based matrices

- **Key issue to design a cement-based matrix for waste immobilization : limitation of adverse cement-waste interactions which may decrease the quality of the final product**

  ➡ Selection of 4 cements to cover a wide range of pH conditions



- **Design of self-levelling mortars with 10% wt. getter** *(Farcy, 2020; Lanier, 2020)*
  - Limited influence of the getter on the cement hydration process : slight acceleration (MKP) or retardation (CSA, CEM I, CEM V) of cement setting, no influence at later age
  - Chemical evolution of the getter in the cement matrix:
    - Destabilization of Ag_2CO_3 at the expense of Ag_3PO_4 (MKP) or Ag_2O (CEM I, CEM V, CSA)
    - Sorption of ions released by dissolution of cement anhydrous phases onto γ-MnO_2



MKP matrix immobilizing metallic waste

➡ **Influence on the trapping properties of the getter ?**

Development of specific packages - Investigation of H_2 sequestration by the cement-based matrices

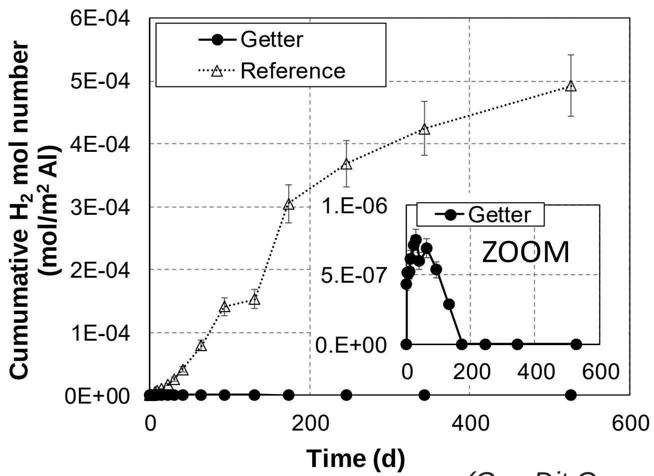
- ***In situ* production of H_2, used as a surrogate of HT/T_2, in the cement matrices by:** *(Farcy, 2020; Lanier, 2020)*
  - External gamma-irradiation : H_2 produced by **radiolysis** of water
  - Encapsulation of Al metal: H_2 produced by metal **corrosion** ($Al + 2\ OH^- \rightarrow AlO_2^- + ½\ H_2$)

➡ Measurement of H_2 outgassing by the cement matrices produced with 0 (getter replaced by inert filler of same particle size) or 10% getter

- **Example: H_2 released by the MKP matrix encapsulating Al powder (8.7 m² of Al / L of matrix)**

Strong reduction, by more than 3 orders of magnitude, of the outgassing

The getter still works in the cement matrix



(Cau Dit Coumes, 2021)

Prospects (PIA PACH³)

- Quantify the trapping performance (kinetics, total capacity) of the getter in the different cementitious environments
- Validate the results with tritiated waste

PIA MACH³

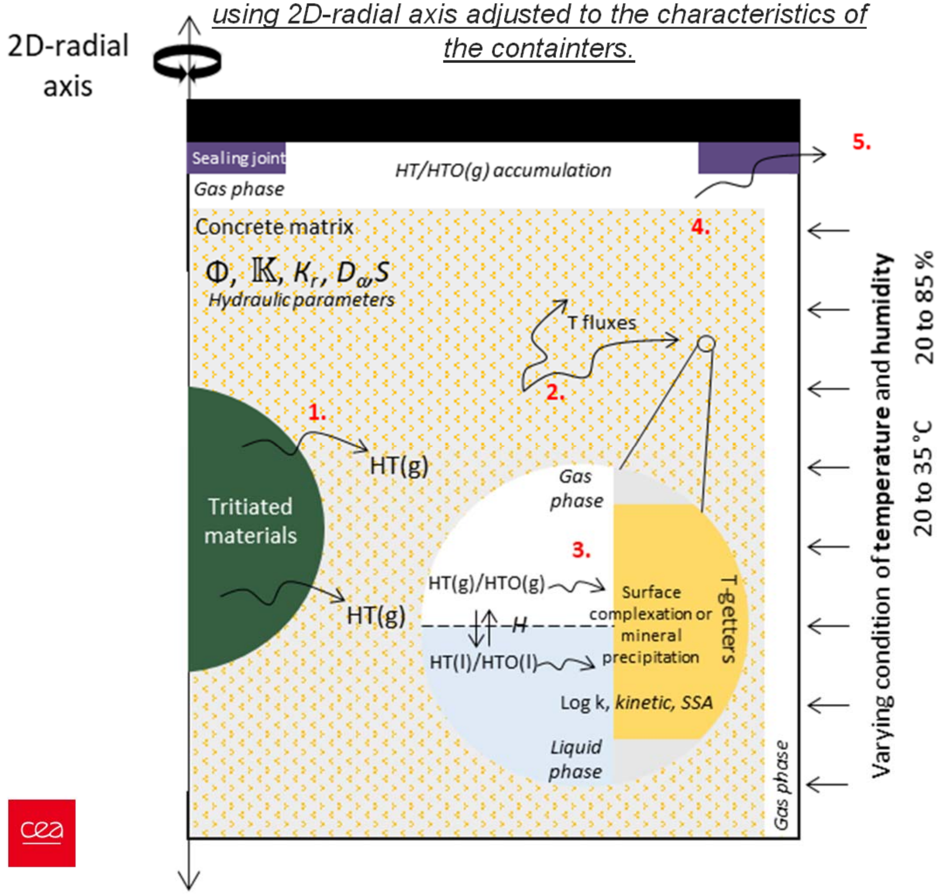
Performance assessment: modeling approach

Development of specific packages- Modeling strategy

- **Overall modeling of the T transfer in the taking into account of the getter**

Multiphasic reactive transport modeling approach using numerical tool HYTEC (van der Lee et al., 2003; Sin et al., 2017)

Schematic representation of the modeling strategy using 2D-radial axis adjusted to the characteristics of the containters.



Chain of chemical and hydrodynamic processes used to simulate Tritium behavior in the concrete matrix:

1. Tritium emission considering a **source term** based on total T content and radioactive decay,

2. Tritium transfer through concrete depending on **hydraulic parameters** (porosity, intrinsic and relative permeability, desorption isotherm and diffusion coefficient) and **radioactive decay**,

3. Tritium contact with tritium-getters. Tritium complexation and/or precipitation,

4. Tritium evacuation from concrete corresponding to the tritium available for sealing joint diffusion,

5. Tritium diffusion through sealing joint. Tritium considered as emitted from the confining package.

Development of specific packages- Modelling strategy

- **Overall modeling of the T transfer taking into account of the getter**

  → Predict T speciation inside the concrete matrix and T outgassing from the concrete matrix within various scenarios:

  - Impact of various T total content,
  - Impact of T-getters content,
  - Impact of humidity and temperature,
  - Impact of T condensation in gas phase
  - Impact of unexpected failure (loss of confinement due to cracks or seal failure).

Conclusion

- **Metallic liner could be part of the solution but not alone**
- **Joint development needed in particular to demonstrate their behavior over long duration**
- **For robustness demonstration, Tritium trapping in the cement matrix may be required (as alternative or complement to the metallic liner)**
  - The trapping getter is selected, with performance evaluation using Hydrogen as simulant
  - But its performance in cement with tritium has to be demonstrated
    - By experiments
    - And coupled to modelling in order to extrapolate the demonstration for long period of time (decades)
- **Need of an overall modelling to quantify the performance of package towards T degassing**
  - Under normal conditions
  - Under abnormal conditions (cracks, seal failure, increase of temperature…)

General references

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