Joint ICTP-IAEA International School on Nuclear Waste Vitrification

23 – 27 September 2019 – Trieste - Italy

DE LA RECHERCHE À L'INDUSTRIE

Overview on French Vitrification Processes

September 24, 2019

Christian LADIRAT

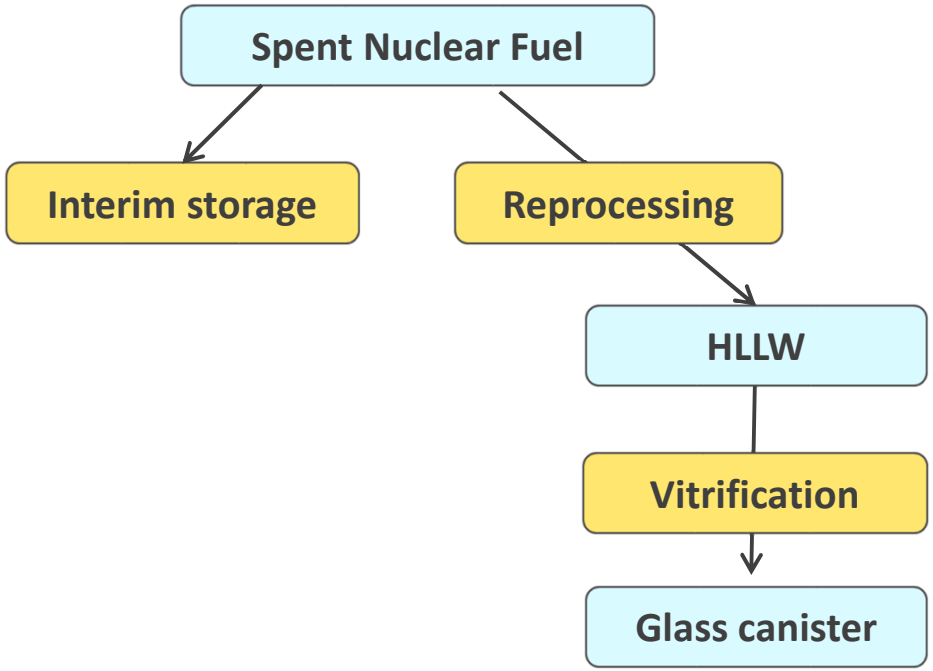
Summary

- History of French vitrification
- Current French Industrial Process
- Cold Crucible Inductive Melter
- New Vitrification Processes

Origin of the High Level Liquid Waste

High Level Liquid Waste (HLLW) comes from spent nuclear fuel reprocessing

- **Defense fuels (Graphite-Gas Reactors) (USA, France, …)**
- **U, Pu Commercial recycling (France, England, Japan, …)**
- **Research Reactors : UMo, SiCrAl,… (Germany, France, …)**



High Level Liquid Waste

< 1% of the total volume of radioactive waste produced in France

> 95% of waste radioactivity

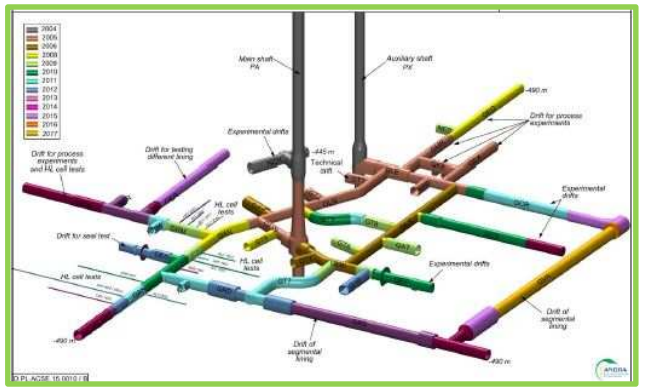
Vitrification Process

4 functions to ensure

- Evaporation
- Calcination: 300 ° C to 600 ° C
  - Transformation into oxides of most elements by nitrates decomposition
- Vitrification : 1050 ° C to 1150 ° C
  - Reaction of calcinate with glassy raw materials
- Off-gas treatment

Produce a package of vitrified waste

- Intermediate storage on site
- Deep Geological Repository



2 large families on an industrial scale

- The two-step process
  - implemented in France and England
- The one-step process:
  - implemented in other countries

History of French vitrification development

Historical overview - First radioactive facilities

Glassy material

Hot Lab

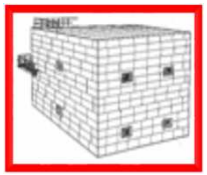
1st VULCAIN (Saclay)

2nd VULCAIN (Fontenay aux Roses)

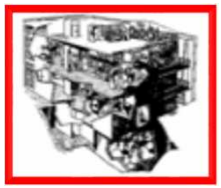
50's 1959 63 66 69

Choice of borosilicate glass

GULLIVER



Induction Heated Metallic Melter PIVER

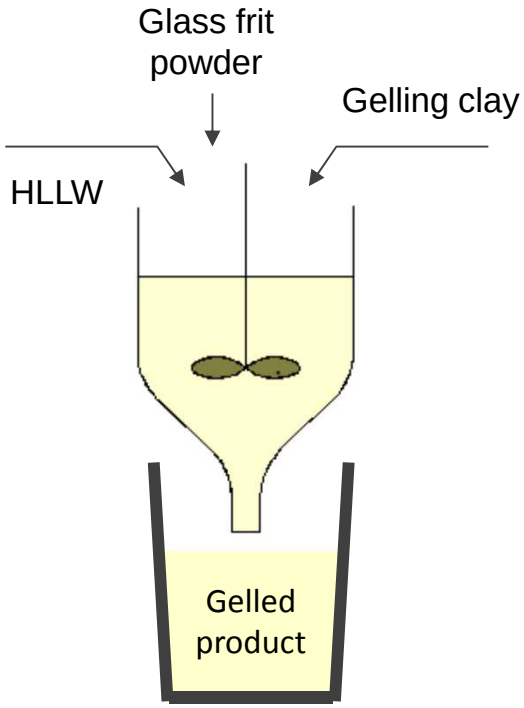


Industrial scale

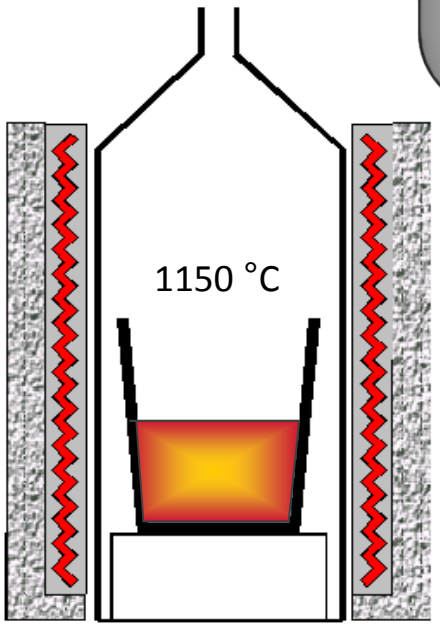
GULLIVER Process (1963-1966)

First achievement of nuclear waste vitrification

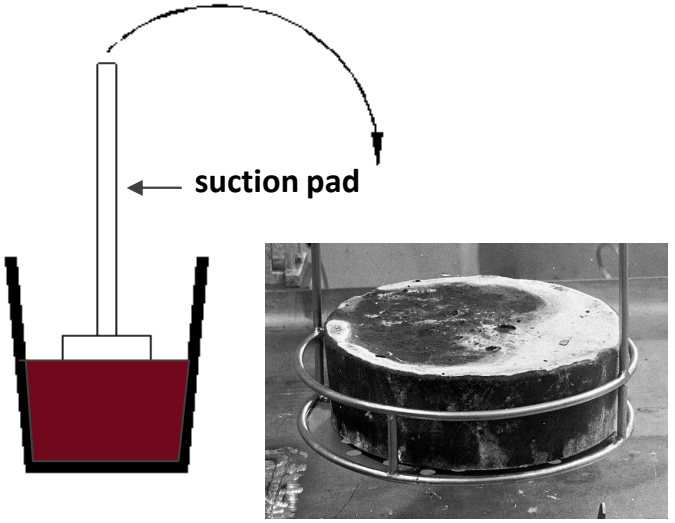
- Batch process in graphite crucible
  - Resistor heated
  - Controlled cooling
  - 4 kg of glass per batch
  - 50 blocks of glass (4 kg each)
  - 120 Tbq per block
  - 250 L of HLLW treated



Jellification

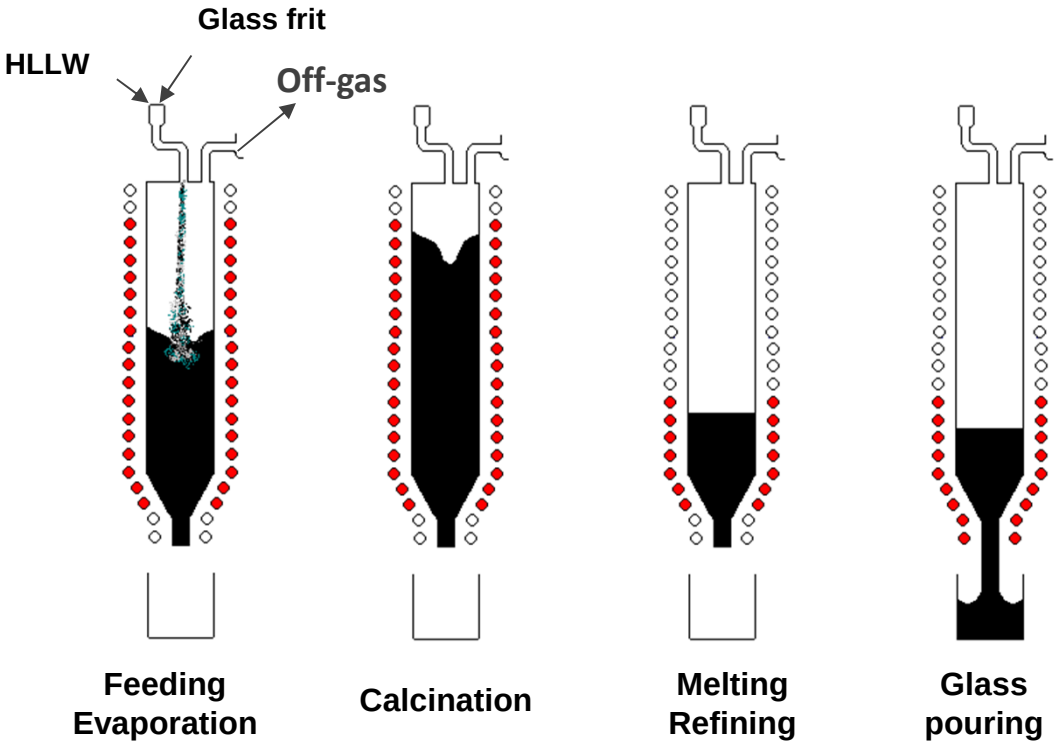


Melting

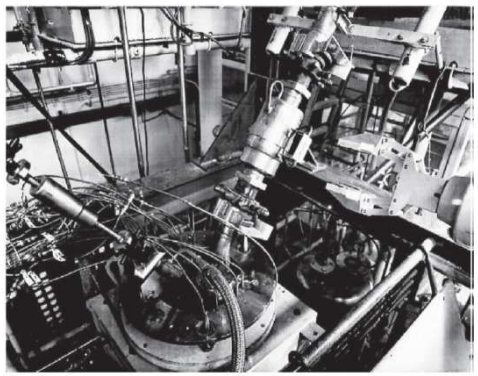


Mold removal

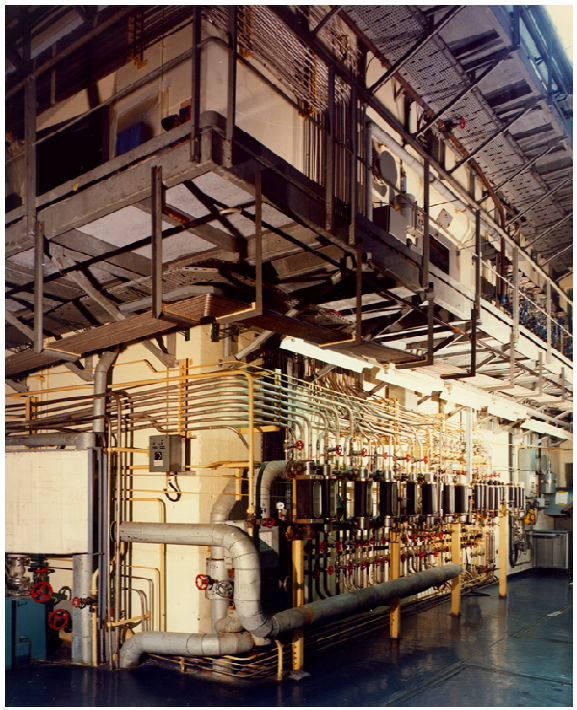
PIVER Process (1969 – 1980)



- Batch process in metallic pot
- Inductive heating (10 kHz)
- Pouring
- 5 kg of glass / h
- 30 m3 of HLLW
- 12 MT of glass
- 164 glass canisters



top view of the metal pot



PIVER hot cell

Current French Industrial Process

An historical overview – Industrial Development

Hot Lab Glassy materials studies

3rd VULCAIN (Marcoule)

2nd VULCAIN (Fontenay aux Roses)

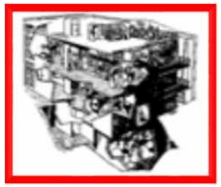
1st VULCAIN (Saclay)

50's | 1959 | 63 | 66 | 69 | 71 | 78 | 89 | 92

Choice of borosilicate glass

GULLIVER

Induction Heated Metallic Melter PIVER



Two-Step Vitrification Process

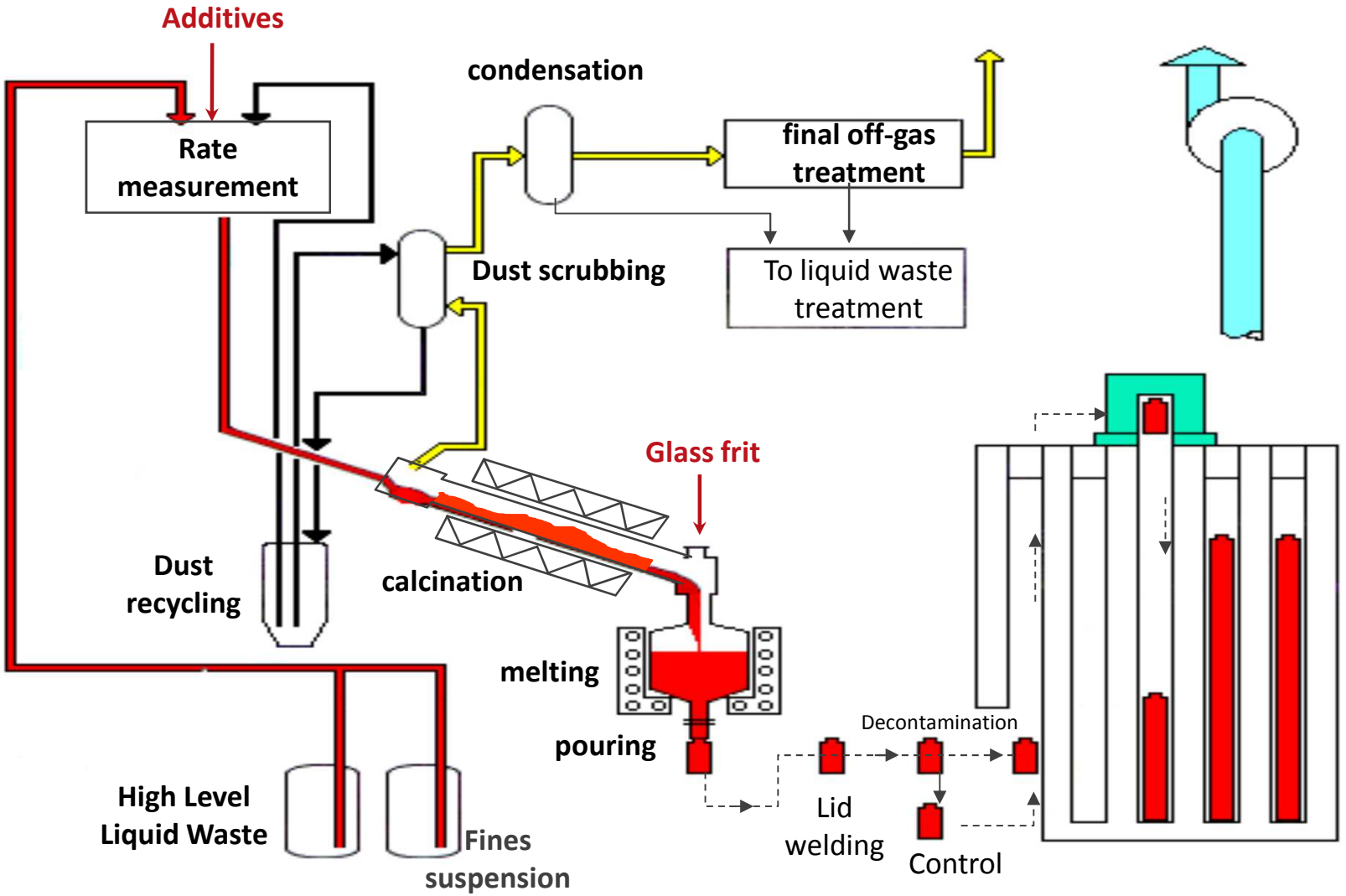
AVM Start up

R7 start up

T7 start up

Industrial facilities

Schematic Diagram of the French Continuous Vitrification Process



Marcoule Vitrification Facility : AVM

Start-up in 1978

Final shutdown in 2012

Calciner : 40 L/h

Metallic Melter : 15kg/h of glass

- Cylindrical shape
- Induction Heated
- Continuous feeding

INDUSTRIAL RECORDS

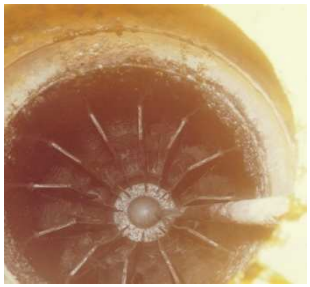
2830 m3 of HLLW treated,

1 220 Mt of glass produced,

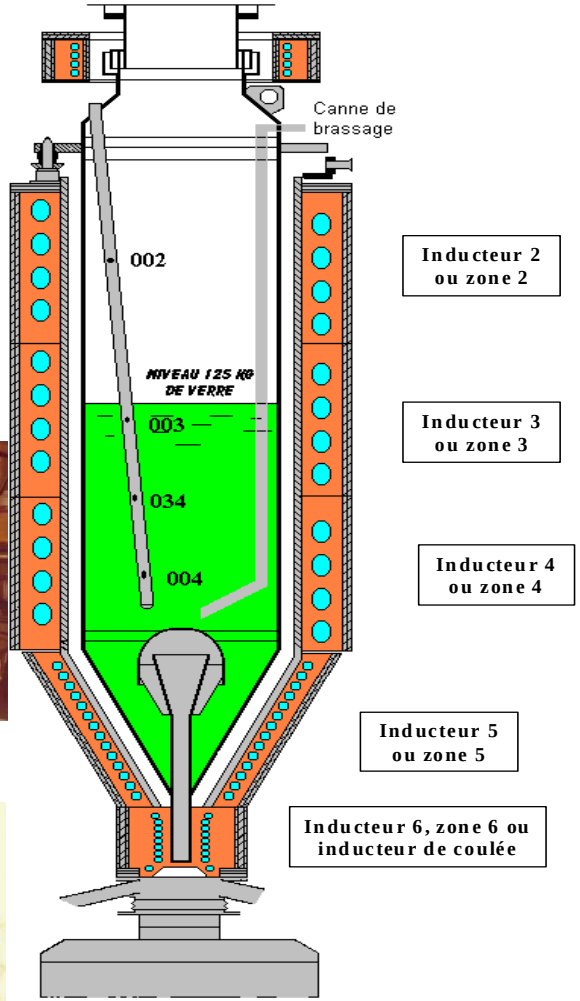
3 306 canisters containing 2210^6 TBq βγ

Cylindrical melter

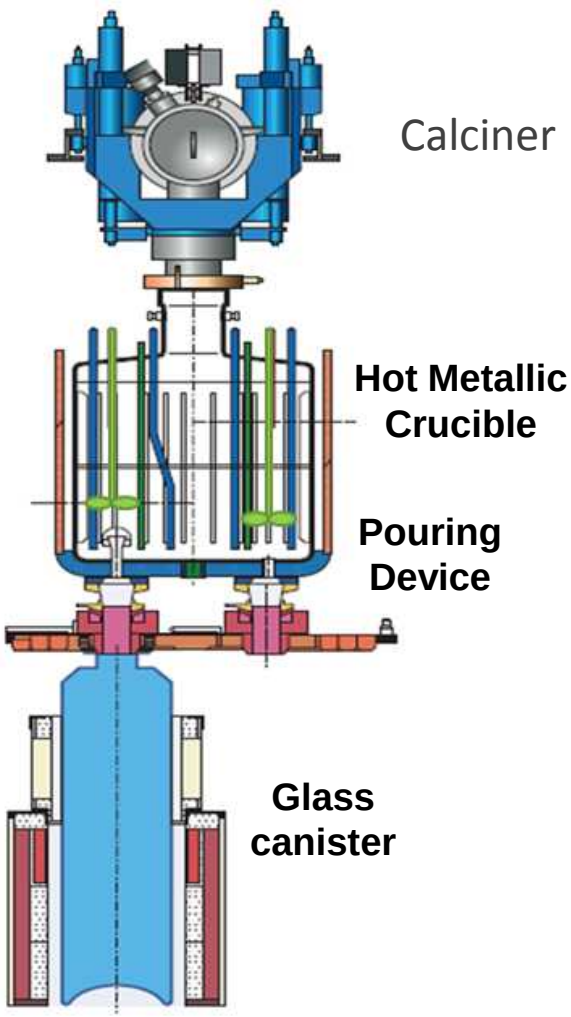
Upper part view



Inside view



La Hague Vitrification Facilities : R7-T7

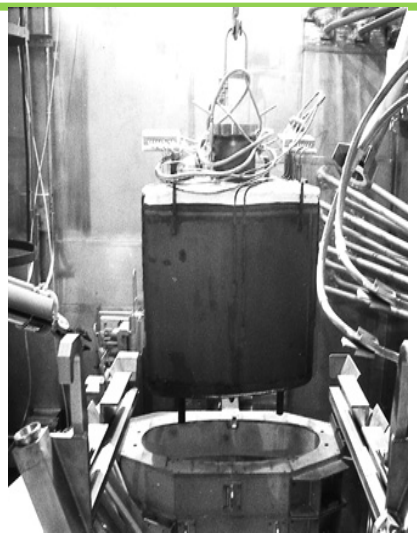


- HLLW coming from Oxide Fuel Reprocessing
- flow-rate up to 90 L/h

- Inductive joule effect into metallic wall
- Thermal flux from metallic walls to molten glass
- Temperature : 1100 °C
- 25 kg of glass / h

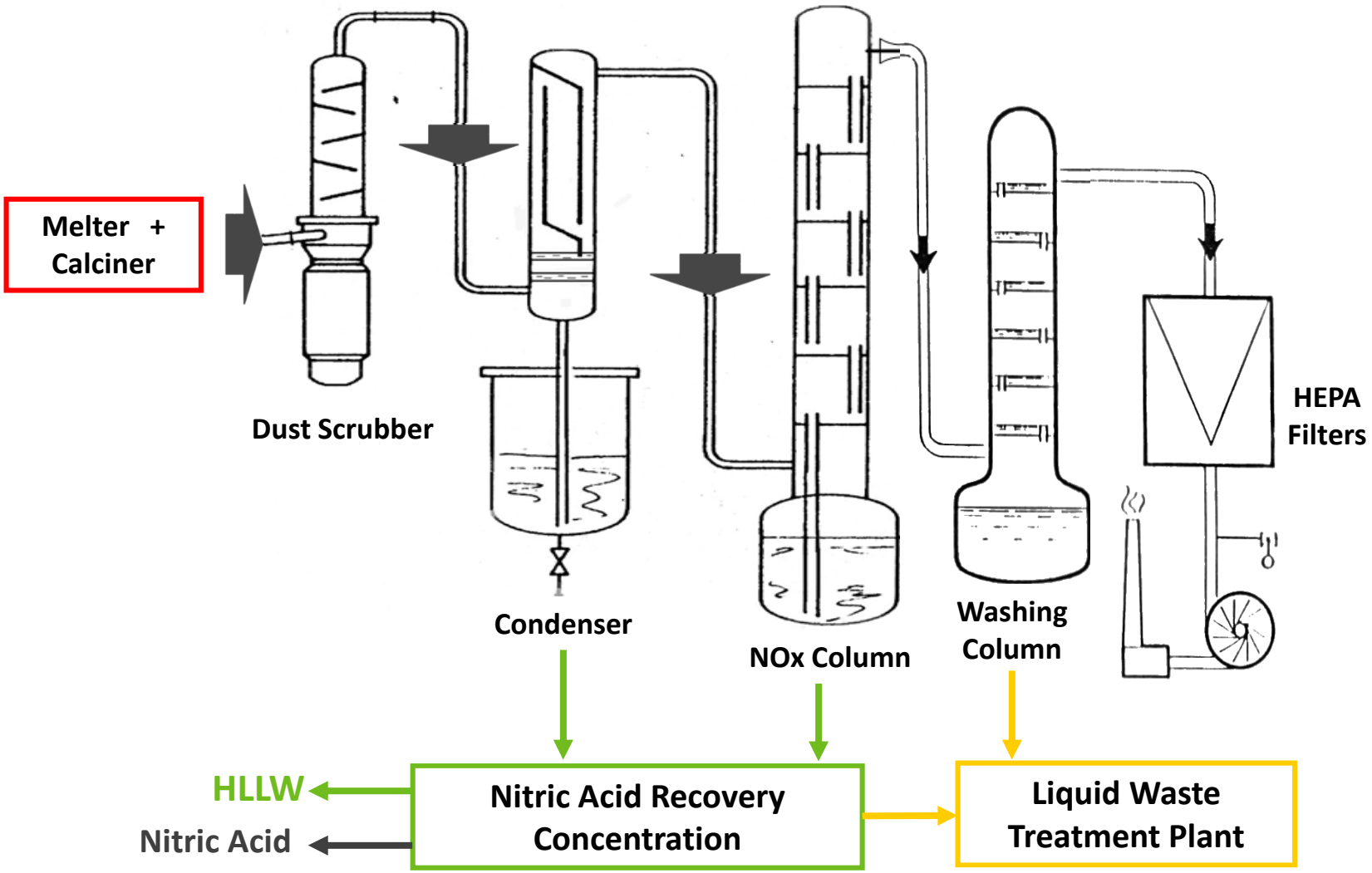
- Upgraded with 4 bubblers and 2 mechanical stirrers
  - up to 3 wt% of Noble Metals

Glass canister



Metallic melter

Off-gas treatment



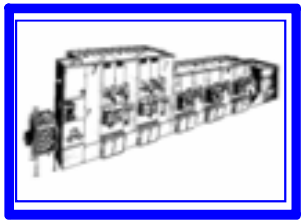
Cold Crucible Induction Melter

An historical overview – R&D support for industrial facilities

Melter development

3rd VULCAIN (Marcoule)

CCIM initial studies



Full scale CCIM

Glassy material

Hot Lab

2nd VULCAIN (Fontenay aux Roses)

1st VULCAIN (Saclay)

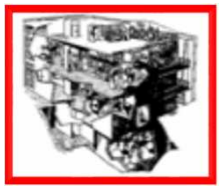
DHA (Marcoule)

50's 1959 63 66 69 71 78 85 89 92 2000 05 10

Choice of borosilicate glass

GULLIVER

Induction Heated Metallic Melter PIVER



Two-Step Vitrification Process

AVM Start up

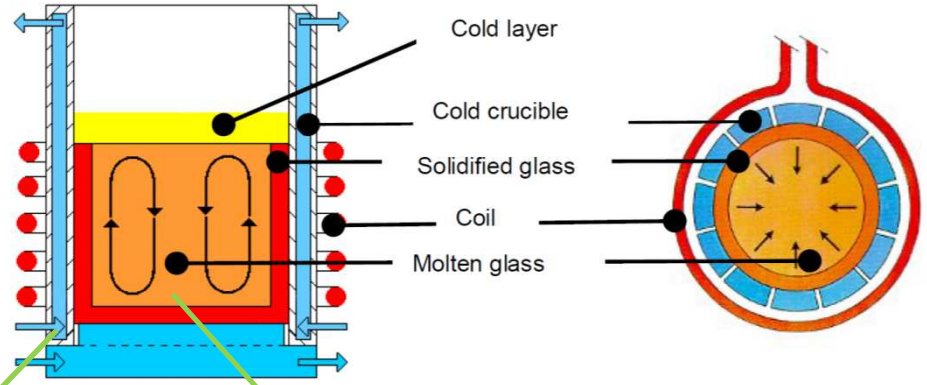
R7 start up T7 start up

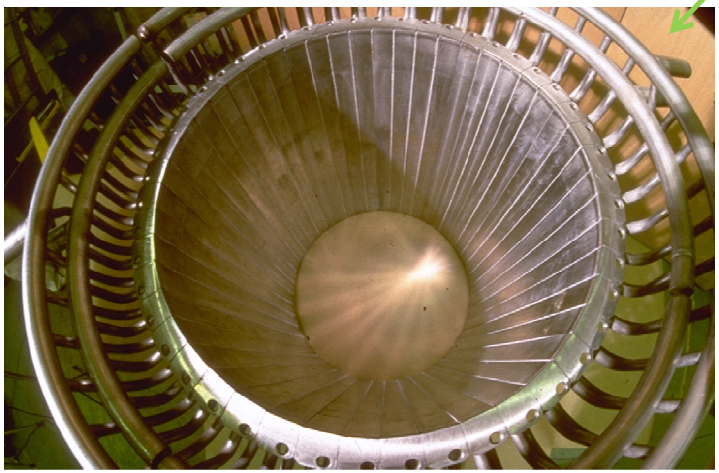
CCIM Start up

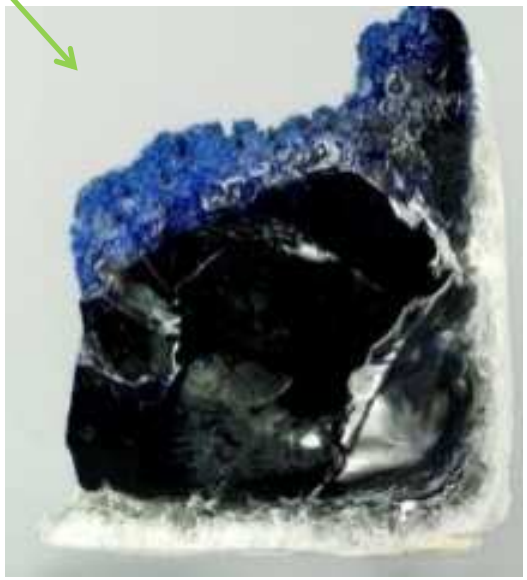
Industrial facilities

Principle of Cold Crucible Induction Melter

- Sectorized structure
- Currents directly induced into the molten glass
- Glass heated by the Joule effect
- Cooled structure ➔ Solidified layer of glass
- T° ~ 1200 °C – 1300°C





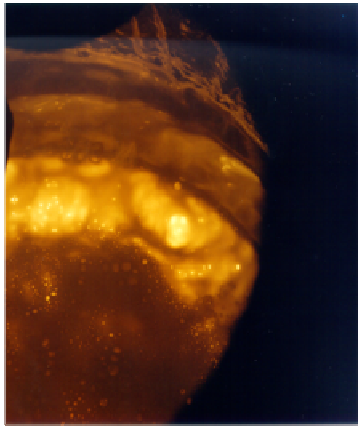


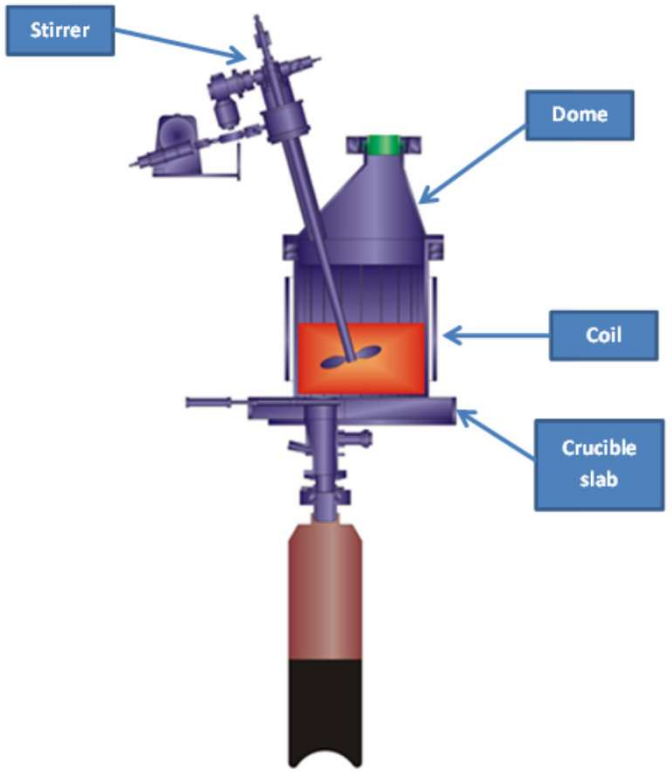
Cold Crucible Induction Melter – Industrial application

- **Legacy Liquid Waste UMo**
- **Intermediate Level Liquid Waste from D&D operation**

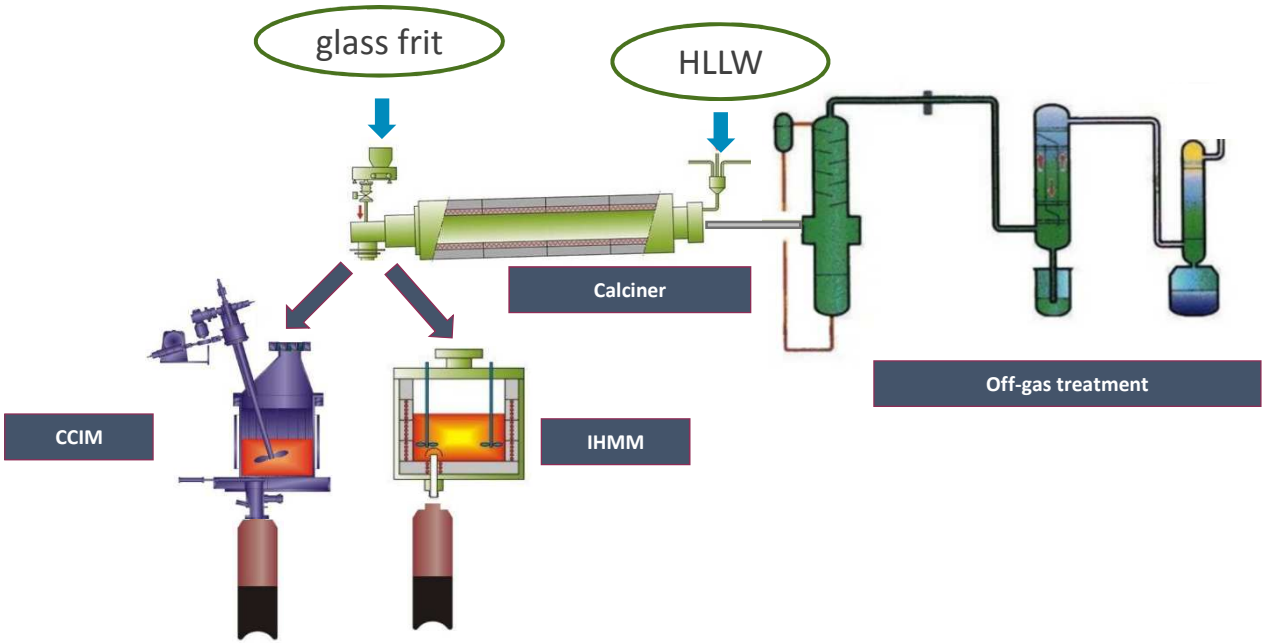
The CCIM is composed of the following elements:

- The metallic crucible shell. Segmented structure transparent to the electromagnetic field.
- The crucible bottom (slab), which includes the pouring valve.
- The dome
- The mechanical stirrer.
- Glass level and temperature specific sensors.
- Bubblers (positioned on the crucible slab).





R7 – T7 La Hague CCIM Vitrification Process



- **Temperature can be increased up to 1300°C**
  - New matrices for D&D effluents with Ce, sulfates, Mo_2Zr,…
  - Waste loading can be increased
  - Glass ceramic with 12% of MoO_3
  - Higher throughput (25 to 36 kg/h)
- **Cold cap layer reduces volatility during feeding including cesium**
- **Corrosion resistant : skull layer protects the crucible from the corrosive melt.**

Major On-line Developments

R & D teams provide continuous support that enhances industrial operation throughout the life of the facilities

- **Extending Melter Service Life (10 years R&D)**
  - major improvements in melter design, material selection, equipment fabrication procedures, and operation procedures.
  - **Melter service life : 5000 hours - 150 Glass canisters - One melter / year / vitrification line**
- **Mechanical Stirring of the Melt (1996)**
  - Greater melter thermal homogeneity → Contribute to extend melter life-time
  - **Higher noble metals content in glass without pouring problems (up to 3 wt%)**
- **Calciner availability**
  - Modification of calciner design
  - Optimization of maintenance operation
  - **Increase of the flow rate from 75l/h to 90l/h**
- **Improvement of Dust Scrubber operation**
  - Design modification (washing flow-rate measurement, modelling,…)
  - Optimization of Dust Scrubber Operation **(better recycling of Cesium into calciner)**
- **Cold Crucible Inductive Melter Implementation (2010)**
  - **Decontamination effluents (Corrosive effluents)**
  - **Highly corrosive liquid waste (UMo reprocessed fuel)**
  - **Future HLLW from reprocessed oxide fuels (higher throughput)**

New vitrification processes

In Can Melter Vitrification Process – One step process

Glassy material

Hot Lab

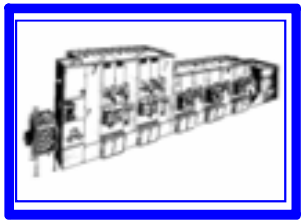
Melter development

3rd VULCAIN (Marcoule)

2nd VULCAIN (Fontenay aux Roses)

1st VULCAIN (Saclay)

CCIM initial studies



DHA (Marcoule)

Full scale CCIM

In Can Melter

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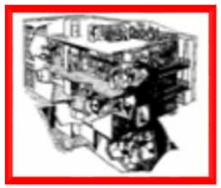


Two-Step Vitrification Process

AVM Start up

R7 start up T7 start up

CCIM Start up

Dem&Melt Melter

Industrial facilities

In Can Melter Vitrification Process

What kinds of waste can be considered ?

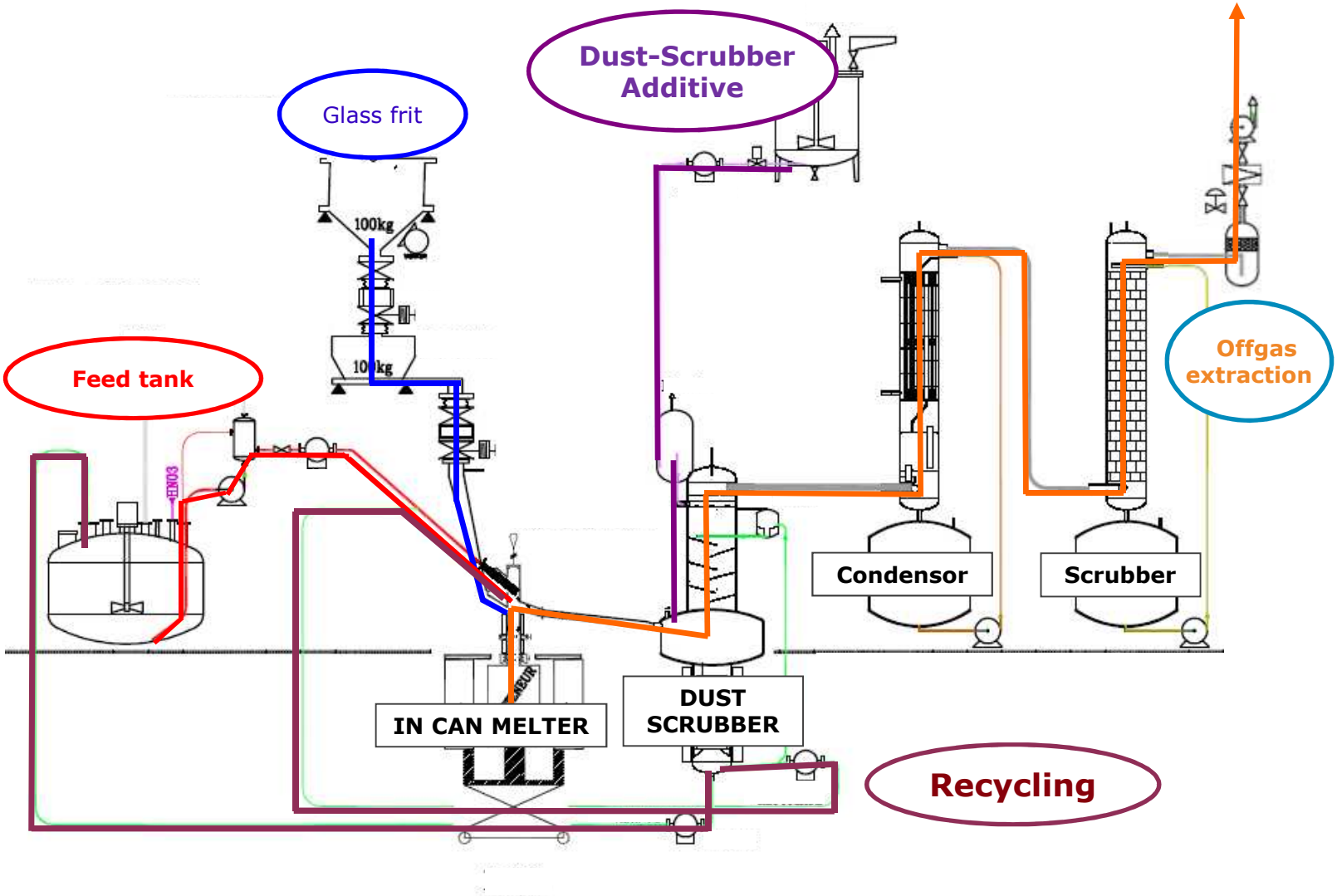
- Alpha bearing liquid waste coming from defense activities
- Liquid Waste coming from dismantling
- Liquid Waste coming from remediation
- Solid waste coming from D&D

What kind of process would be developed ?

- In Can is a simple and robust process
  - Resistance heating
  - Single use Canister in a furnace (no draining)
  - Corrosion resistant (single use crucible)
  - Liquid feeding of the melting pot (1 step, no calciner)

- Some disadvantages
  - Batch process
  - Small treatment capacity
  - Thermal homogenization (no stirrer)

In Can Melter Vitrification Process

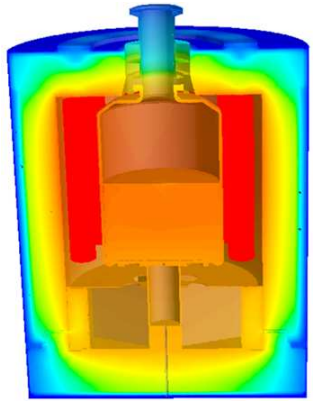


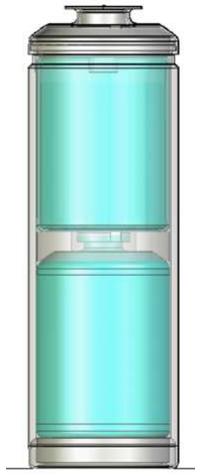
In Can Melter Vitrification Process – First application

Alpha bearing liquid waste at Valduc site

- Low volume of effluents
- Implementation in glove box
- Operation by campaign with two shifts (morning and afternoon) and keeping molten glass overnigh

- adaptation of glass viscosity
- Two cans filled with glass are packaged in a container similar to those of La Hague





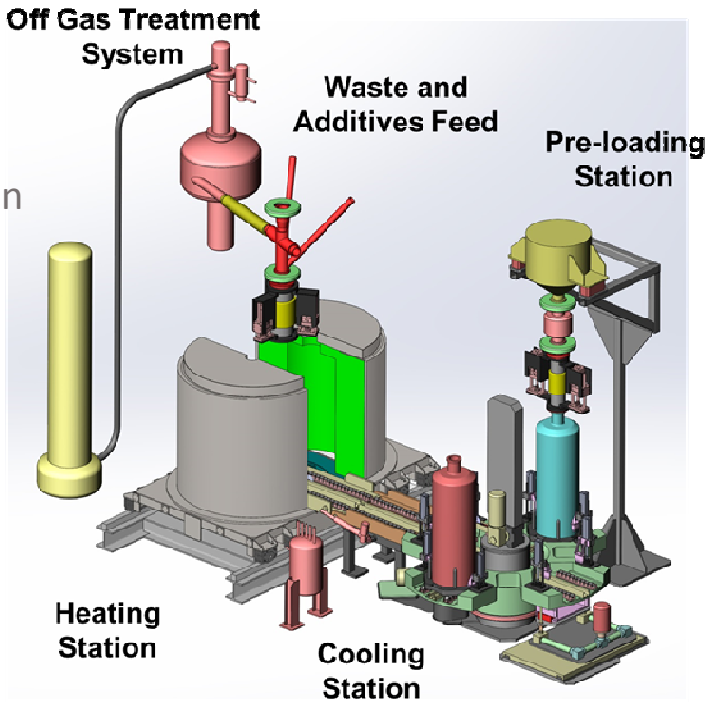
Full scale test rig

This easy-to-use process meets customer needs for a relatively low investment cost

DEM&MELT In-Can Vitrification Process

In-Can vitrification process for solid and liquid waste

- **Targeted waste :**
  - Waste streams **from D&D operations**
  - Waste streams from **remediation operations**
  - **Orphan waste** / Waste with no route
  - Waste identified as an issue in terms of handling, transportation, adapted conditioning matrix or evacuation outlet within the existing regulations



- **Main goals**
  - Volume reduction
  - Durable containment of radionuclides
  - Waste stabilization
  - Must be able to treat solid, liquid, sludge with optional feeding systems
  - Easy to install in an existing dismantling facility
  - Short operating time. Easily dismantled after operation

DEM&MELT In-Can Vitrification Process

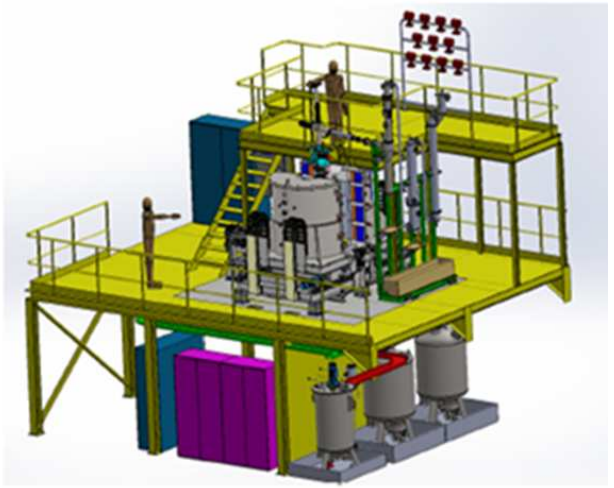
1. Feasibility demonstration for an identified waste from dismantling operations in Marcoule

- Residues highly loaded with Cs (UP1 marcoule)
- Tests performed on liquid and solid form
- Visual and SEM exams highlight a very homogeneous glass matrix
- Only 0,2 %w of the Cs in Off Gas Treatment

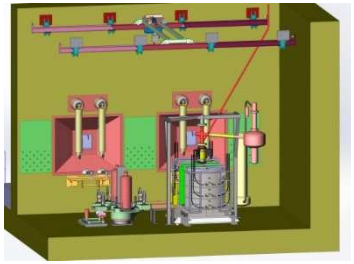
Can cut in half for sampling

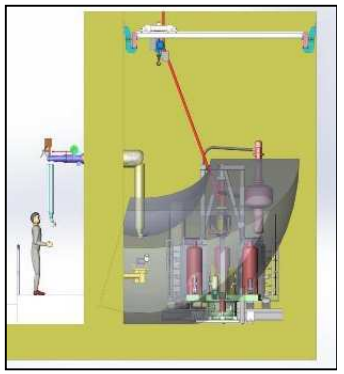
2. Customization of the In Can Vitrification Process for the treatment of nuclear waste resulting from D & D and remediation operations

- Adaptation for Intermediate and High Level Waste
- 300 kg of glass per can
- Feeding system compatible for Solid and Sludge
- Modular system for in situ implementation

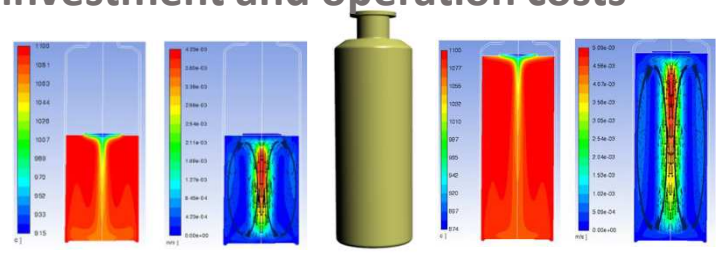


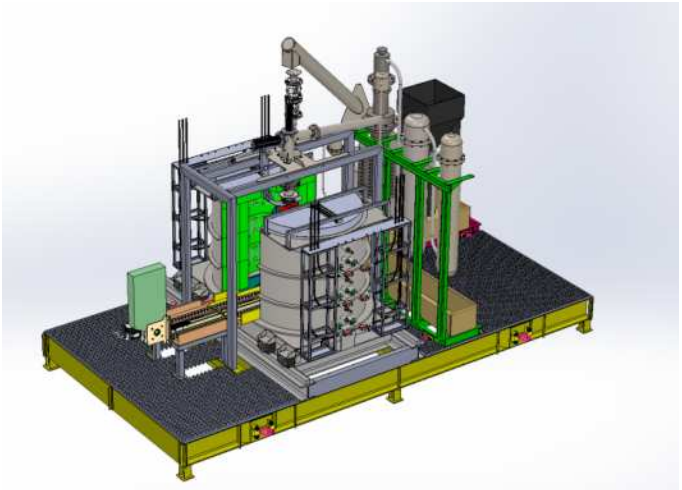
Basic design for nuclear facility





- Canister used as part of the melter (no pouring; no corrosion issue)
- Heated by a simple and **robust resistance furnace**
- Offering a great homogeneity to the melted mixture (no stirrer; no bubbling)
- Suitable for **High or Intermediate Level Waste**
- **Efficient temperature control and Cs volatility management**
- Suitable for **solid or liquid waste**, including zeolites and sludge
- Flexible enough to **accommodate uncertainties on waste composition**
- Designed in order to produce a **small amount of secondary waste**
- **Modular and compact** enough to be implemented in a decommissioned cell or close to the waste to be treated
- Implemented in 20 feet ISO containers
- **Requiring low investment and operation costs**





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Thank you for your attention

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