French HLW vitrification history and major achievements

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Sumglass 2023

Summary

1. French vitrification history
2. La Hague vitrification lines achievements
3. Latest development

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French vitrification history

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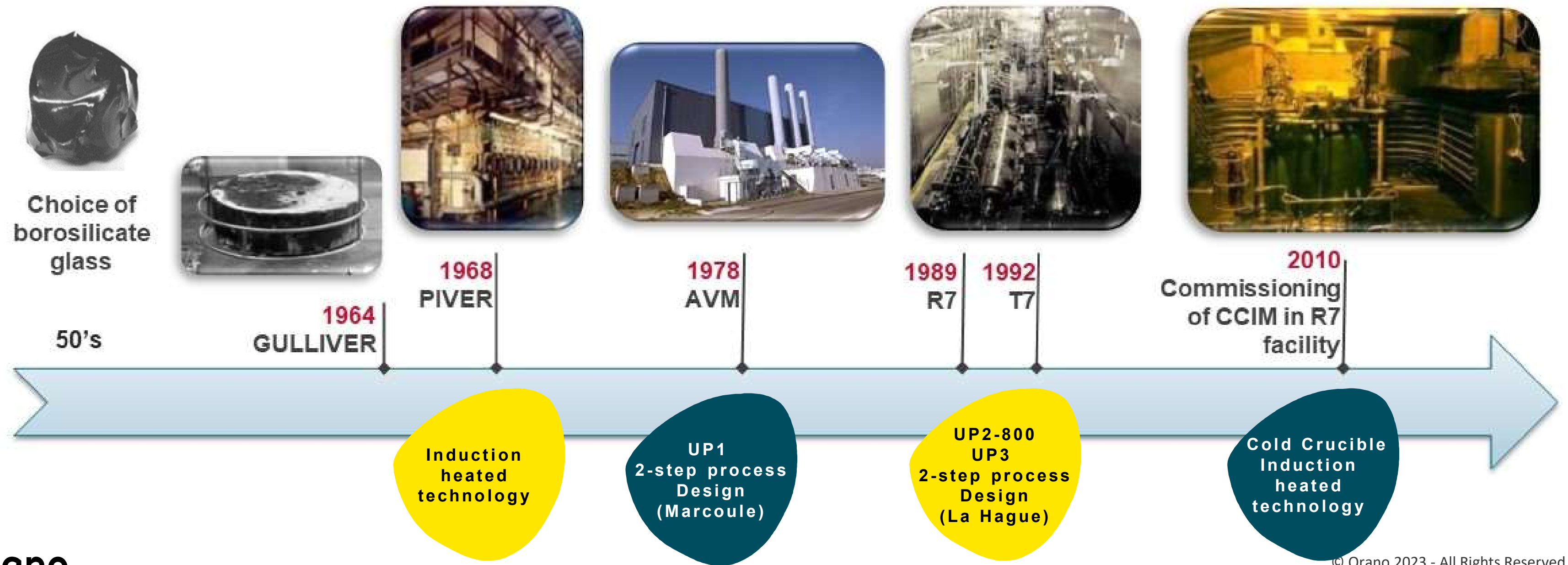
Major French Vitrification Technology Milestones

Vitrification of High-Level Waste (HLW) is the internationally recognized standard to:

- Minimize the final waste volume
- Minimize the impact to the environment resulting from waste disposal



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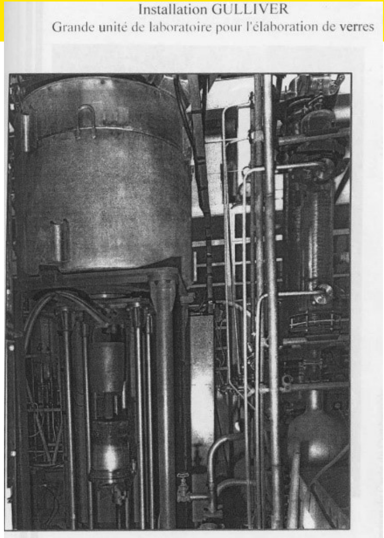
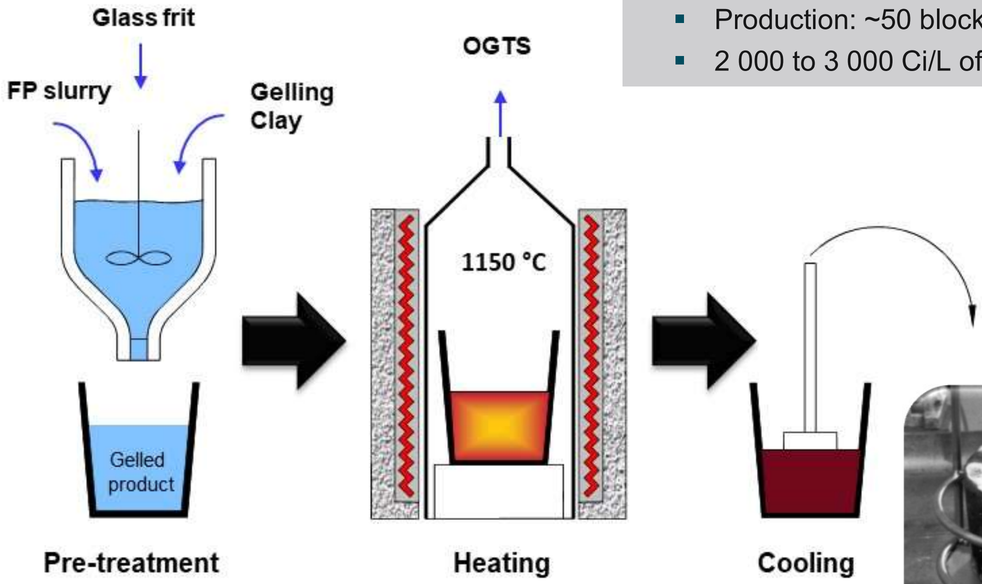
Historical overview
GULLIVER Process (1964 – 1967)

1st Large Scale Active Pilot

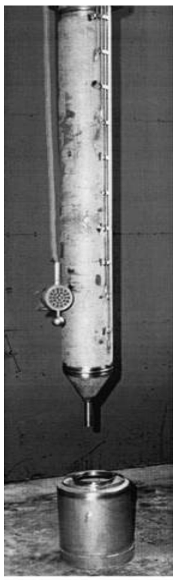
- **Batch process in graphite crucible**
  - Resistance heating
  - Controlled cooling
  - Production: ~50 blocks of 4 kg
  - 2 000 to 3 000 Ci/L of glass

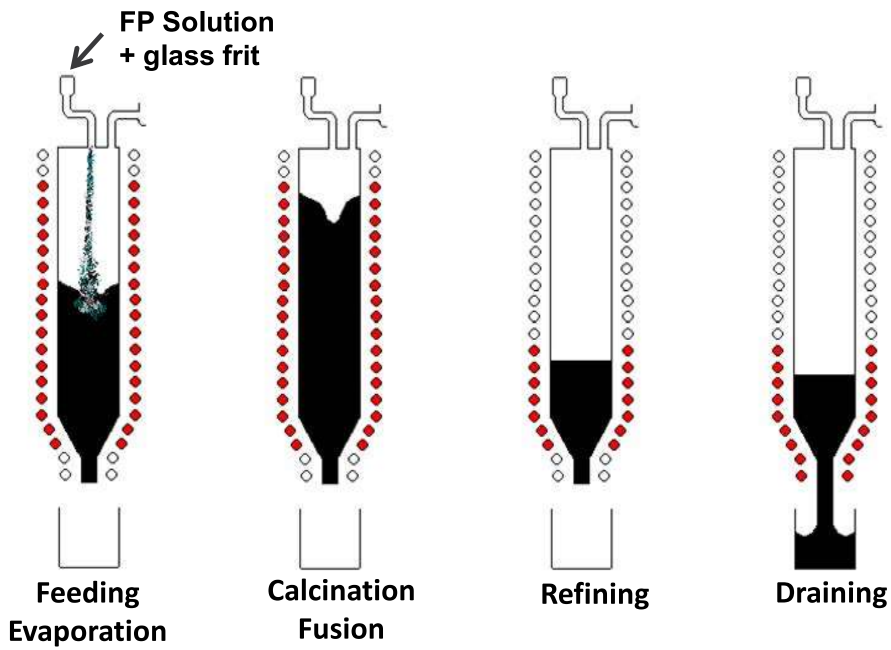




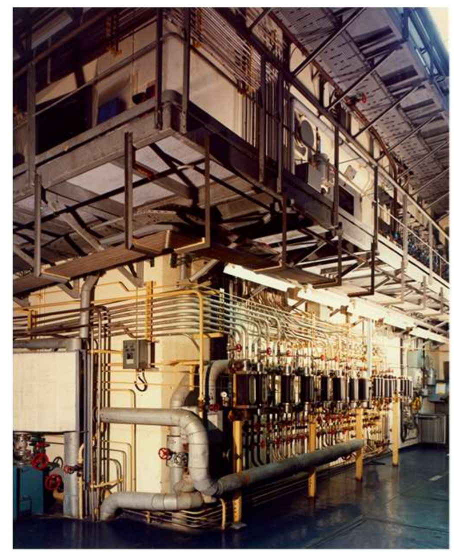
Historical Overview
PIVER Process (1968 – 1980)

1st Industrial Unit





- **Batch process in graphite crucible**
  - Induction heating
  - Draining by the bottom
  - Throughput: 5 kg/h
  - Production: 12 metric tons of glass



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Historical Overview
2-Step Process Reference Design

Over 40 years of Industrial Operation

AVM – Operated by Orano from 1978 to 2009 (Marcoule)

1 vitrification line

- ~ 3 300 canisters produced
- ~ 1 220 metric tons of glass produced
- ~ 22 10^6 TBq vitrified

R7 / T7 – Operated by Orano since 1989 (La Hague plant)

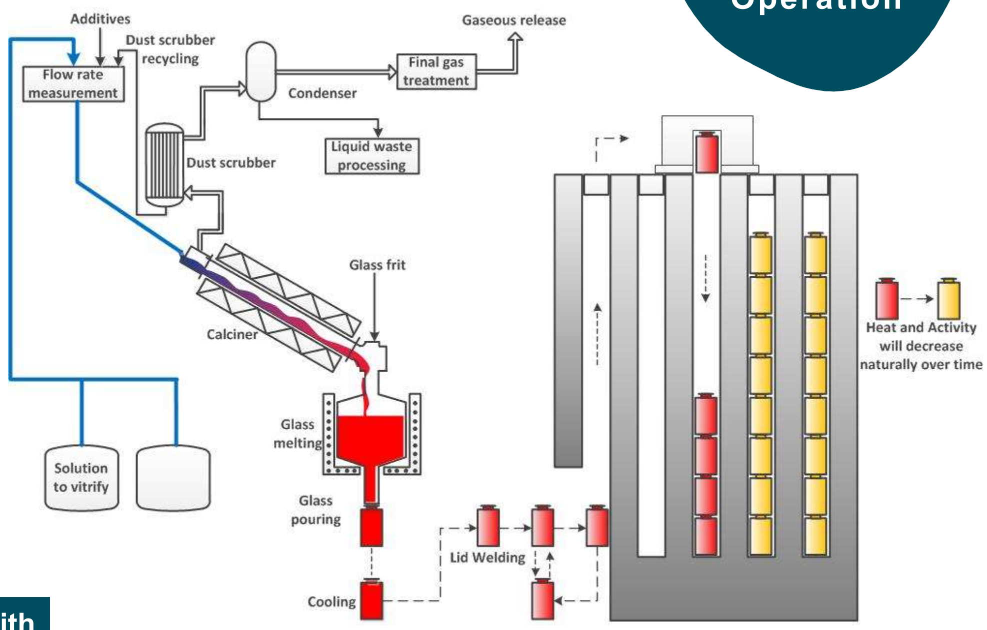
2 vitrification facilities (6 lines)

IHMM (end of 2021)

- ~ 24 100 canisters produced
- ~ 9 600 metric tons of glass produced
- ~ 368 10^6 TBq vitrified

CCIM (end of 2021)

- ~ 1 000 canisters produced



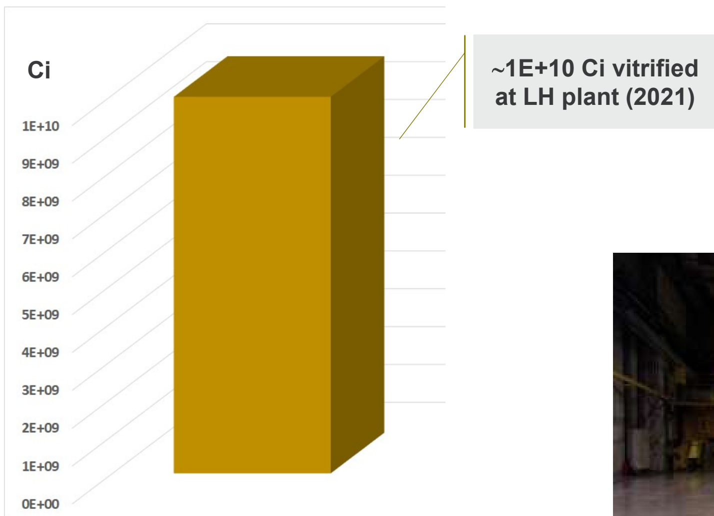
Very High-Level Waste management with significant incorporation rate into the glass

➔ Around 1 Ci/g

Process also implemented at WVP Sellafield

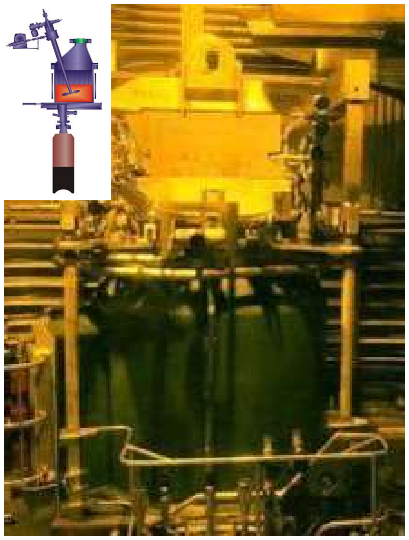
Orano La Hague Vitrification Track Records

Significant amount of radioactivity vitrified over the years



Main vitrification technologies developed

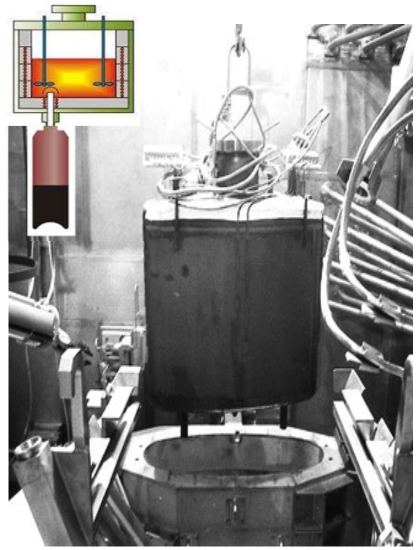
CCIM



Cold Crucible
Direct induction

In operation since 2010

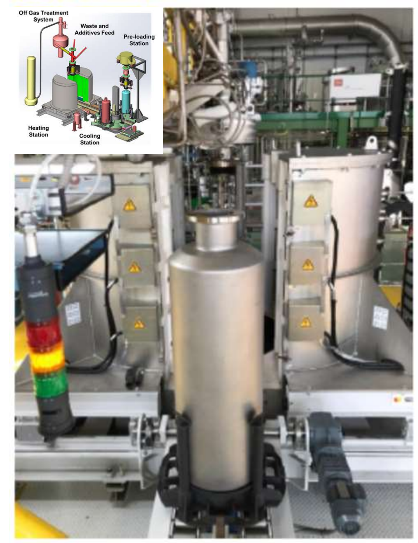
IHMM



Metallic Melter
Indirect induction

In operation since 1978

In-Can Melter



Resistance heating
Thermal homogenization

Full scale pilot commissioned in 2020

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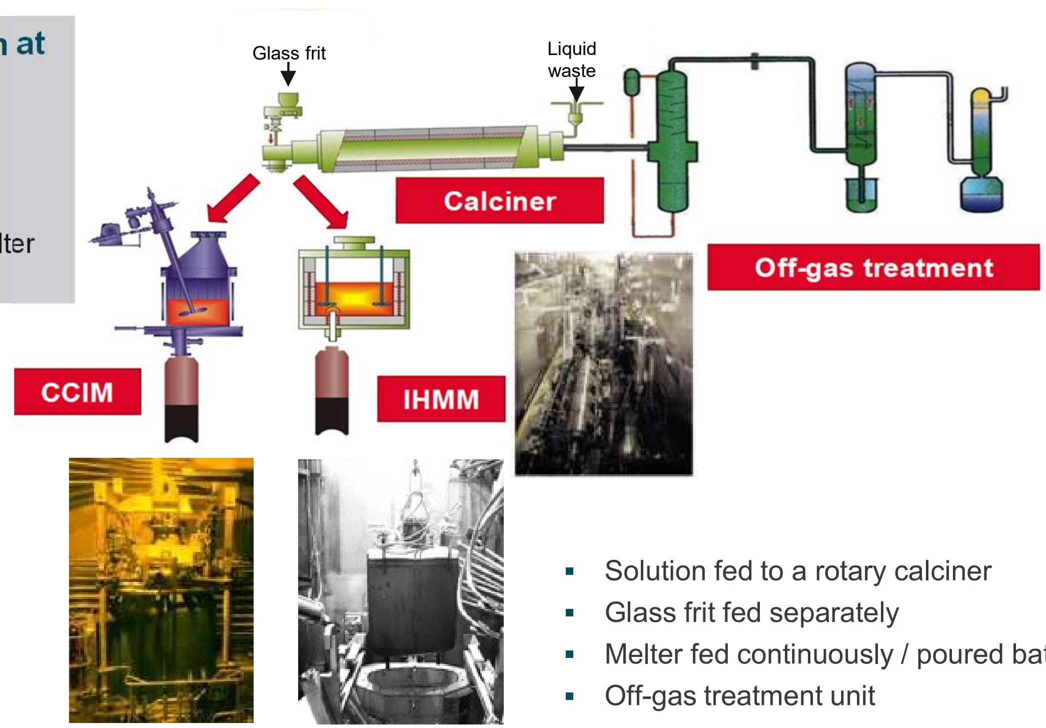
La Hague vitrification lines achievements

R7/T7 Vitrification 2-Step Process

- **2 vitrification facilities in operation at La Hague plant**
  - Up to 1 100 canisters/year
  - 5 lines with **I**nduction **H**eated **M**etallic **M**elter (6 lines from 1989 to 2008)
  - 1 line with **C**old **C**rucible **I**nduction **M**elter (commissioned in 2010)



- Solution fed to a rotary calciner
- Glass frit fed separately
- Melter fed continuously / poured batchwise
- Off-gas treatment unit

R7/T7 Continuous Improvement Policy

- Implementation of several improvement programs since R7 & T7 commissioning
- Major improvements/achievements:
  - ↗ Calciner treatment capacity from 60 L/h to 105 L/h
  - ↗ Calciner reliability and lifetime
  - ↗ Noble Metals incorporation & glass throughtput
  - ↗ Melter (IHMM) lifetime by 400 %
  - ↗ Overall reliability and remote in-cell maintainability
  - Implementation of a CCIM in R7
  - Adaptation of glass formulation for effluents to be vitrified

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R7/T7 Continuous Improvement Policy

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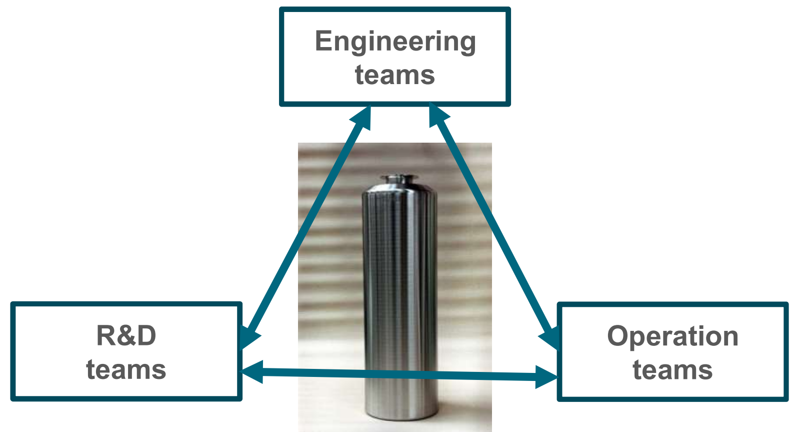
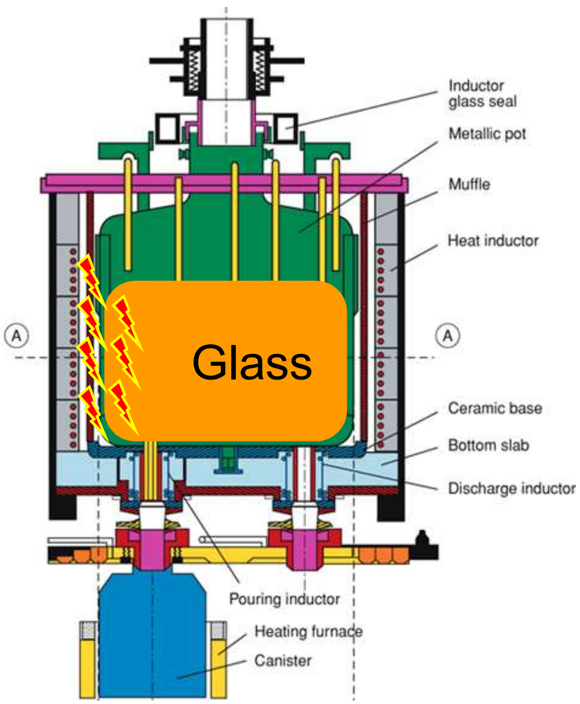
Design improvements

Process parameters optimization

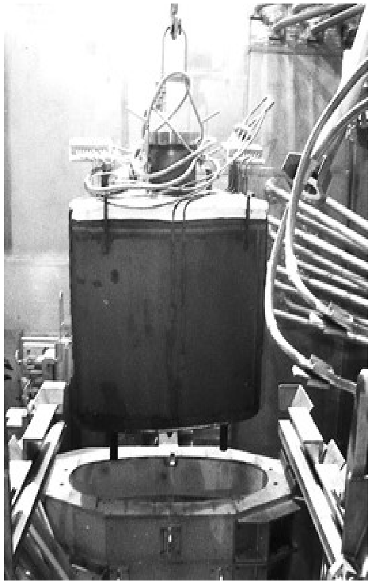
With respect to safety & glass quality



Induction Heated Metallic Melter (IHMM)



IHMM principles



IHMM in hot cell

Design Principles
- Inductive joule effect into metallic wall
- Thermal flux from metallic wall to molten glass
- Mixing ensured by bubbling and stirring

Process operation
- T° ~ 1100 °C
- Calcine fed
- Continuous feeding / Batchwise pouring

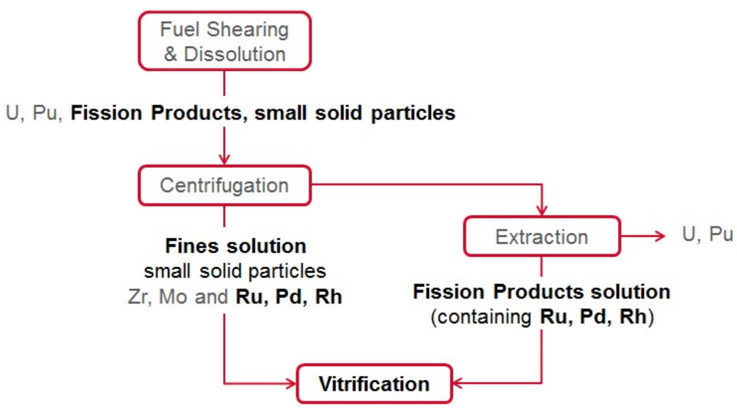
TRL 9
- Over 40 years of industrial operation
- Over 10 800 metric tons of glass produced

Wasteform
- Homogeneous borosilicate glass
- Around 1 Ci/g

Noble Metals Incorporation Challenges

Noble Metals Origin

Ruthenium, palladium and rhodium are part of fission products resulting from nuclear fuel reprocessing



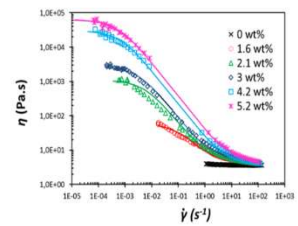
Schematic representation of the La Hague spent fuel reprocessing plant

Noble Metals Challenges

- NM have Low solubility in the glass
- NM modify physical properties of the glass such as electrical and thermal conductivity and viscosity
- NM tend to settle to the bottom of the melter due to their high density
- NM can reduce the kinetics of the glass forming reaction

Potential issues

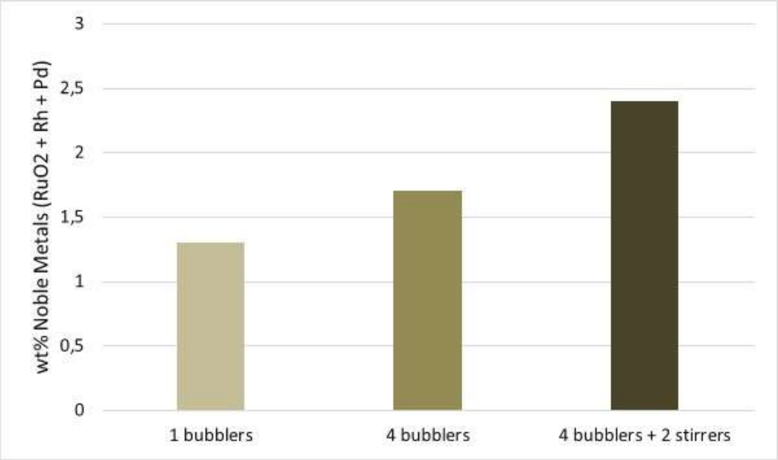
- Pouring problems
- Disruption of heating power distribution in the molten glass
- Risk of short-circuit (electrodes heating systems)
- Decrease of process throughput
- Increase of melter downtimes



All high-level waste vitrification facilities have experienced operational problems arising from the presence of noble metals in the melter

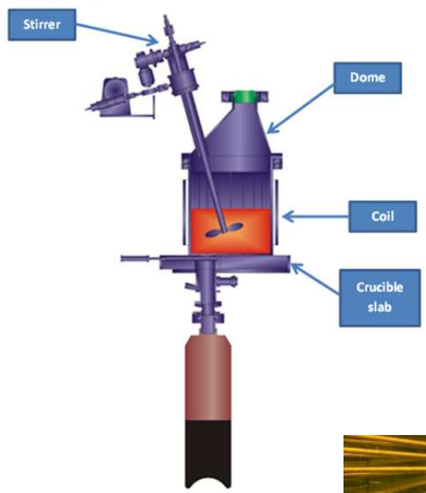
IHMM Noble Metals Incorporation Improvement

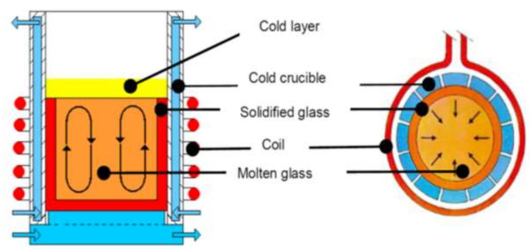
- **Several technological enhancements implemented to the melter design to improve mixing capability**
  - Initially ➔ R7/T7 melters equipped with a single bubbler
  - 1990 ➔ R7/T7 melters equipped with 4 bubblers
  - 1996 ➔ R7/T7 melters equipped with 4 bubblers and mechanical stirring
- **2008-2014 ➔ Optimization program to increase NM incorporation as well as glass throughput**
  - Analyze of industrial operation feedback
  - Identification of key process parameters affecting NM incorporation efficiency
  - Definition of new recommended operating parameters
  - Progressive implementation in the R7 & T7 facilities



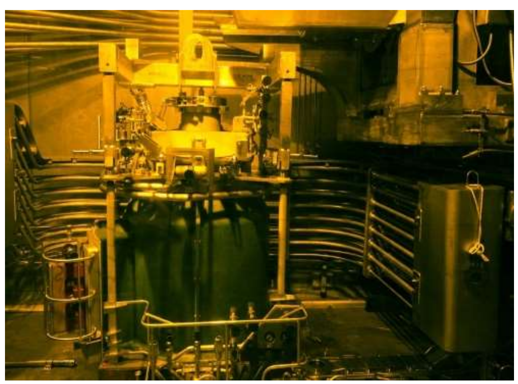
- Enhancing thermal homogeneity
- Enhancing mixing of the glass melt (providing enough shearing to maintain the particles in suspension)
- Better spread of the glass frit and calcine fed to the melter
- Enhancing temperature management
- Improving operating modes

Cold Crucible Induction Melter (CCIM)





CCIM principles



CCIM in hot cell

Design Principles

- Glass heated by Joule effect (Currents directly induced inside the molten glass)
- Cooled structures ➔ Solidified layer of glass protecting the melter from the corrosive melt
- Mixing ensured by bubbling and stirring

Process operation

- T°: beyond 1300°C
- Solid or liquid fed
- Continuous feeding / Batchwise pouring
- High glass throughput reachable (higher T°)

TRL 9

- Over 12 years of industrial operation

Wasteform

- Homogeneous borosilicate glass
- Glass-ceramic
- High waste loading reachable (higher T°)

CCIM deployment in R7 facility

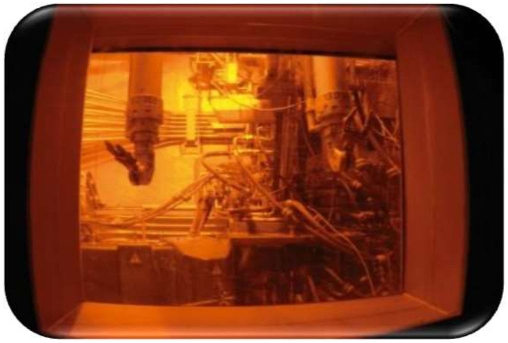
- **2010 ➔ CCIM Commissioning in R7 facility - Line B**
  - **Vitrification of effluents from D&D operations**
  - ➔ Requiring high T° to reach significant WL
  - **Vitrification of legacy FP from reprocessing of U-Mo-Sn-Al spent fuel**
  - ➔ High content of Mo & P making the glass extremely corrosive
  - ➔ Requiring high T° to reach significant WL

D&D glass

UMo glass-ceramic

Implemented in a very harsh environment with respect to challenging constraints

- Installed in place of an IHMM without impacting the existing structures and upstream or downstream equipment
- Installed in a highly radioactive cell into which human access was prohibited (whole work carried out remotely)
- Installed while lines A and C were in operation



CCIM Operation feedback

- **Production**
  - Vitrification of effluents from the La Hague UP2-400 facility D&D operations ➔ ~220 canisters
  - Vitrification of the whole inventory of the UMo legacy FP ➔ ~750 canisters
  - Throughput achieved: ~ 135 kg/h/m^2
- **Improvements implemented since commissioning**
  - ➚ bubblers lifetime
  - T° probes reliability enhancement (patent US8182146)
  - Mechanical stirrer optimization
- **Current status**
  - Vitrification of the very high-level fission products coming from the ongoing reprocessing activities at La Hague Plant
  - Average incorporation rate of 1 Ci/g of glass
  - Glass loaded with up to 3 wt% of NM (RuO_2 + Rh + Pd).

This achievement is the outcome of the continuous improvement policy implemented through a close collaboration between R&D, engineering teams and the industrial operator

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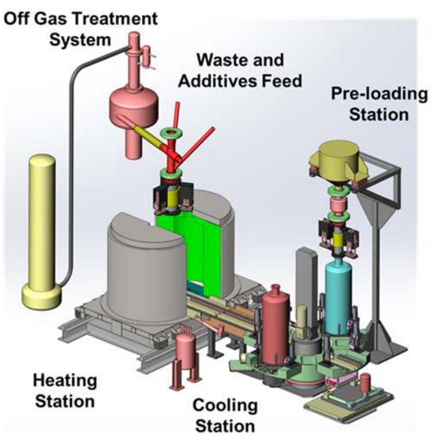
Latest development

DEM&MELT In-Can Melter

Suitable for ILW and HLW



DEM&MELT principles

DEM&MELT full-scale pilot

Design Principles

- Electrical resistance heating
- Canister used as the melter (no pouring device)
- Mixing ensured by heat convection
- Scalable

Process operation

- Operating temperature range ~ 100°C - 1150°C
- Solid or liquid fed
- Batch process

TRL 7

- Full scale pilot commissioned in 2020
- Design benefiting from proven technologies

Wasteform

- Homogeneous borosilicate glass
- Composite matrices
- Waste encapsulation
- High waste loading reachable (up to 80 wt%)

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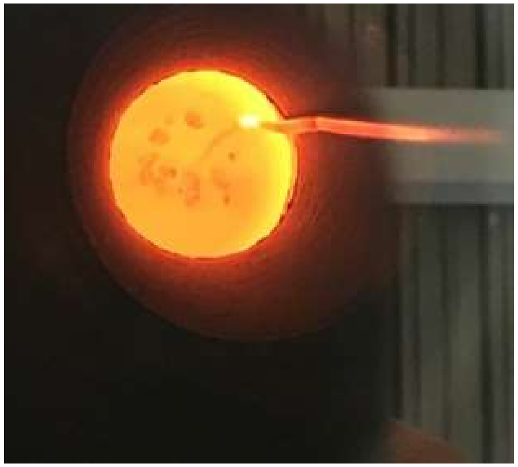
State of Development

DEM&MELT full scale pilot commissioned in November 2020

► Proper operation on the whole perimeter: mechanical system, feeding system, melter, off-gas treatment system



Molten materials surface as observed during the DEM&MELT start-up test

State of Development

DEM&MELT full scale pilot commissioned in November 2020

- Proper operation on the whole perimeter: mechanical system, feeding system, melter, off-gas treatment system

Mature and proven technologies *from up stream functions to down stream functions with over* ***40 years of HA operation*** *at Orano La Hague and CEA Marcoule*

- **Remotely-controlled operation and maintenance functions**
  TRL 9 – *used in R7/T7 and ACC*
- **Canister management functions**
  TRL 9 – *used in R7/T7 and ACC*
- **Off-gas treatment system**
  TRL 9 – *used in R7/T7*
- **Feeding systems**
  TRL 9 – *used in R7/T7*
- **Auxiliary functions**
  TRL 9 – *used in La Hague and Marcoule*

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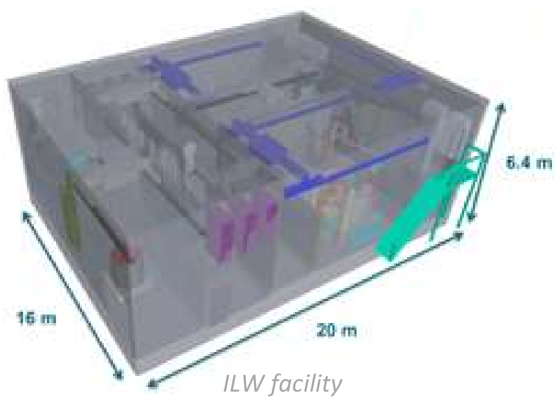
Industrial design

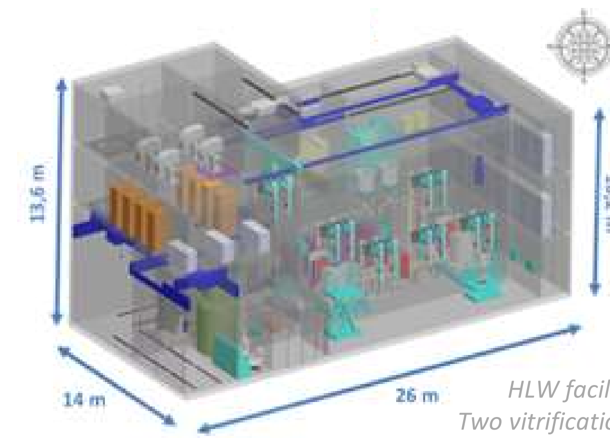
Design Requirements

- Simple and compact equipment
- Modular design
- ↓ Investment and operation costs
- ↑ Maximize availability
- ↓ Occupational exposure and environmental impact
- ↓ Secondary waste



ILW facility
Two vitrification lines

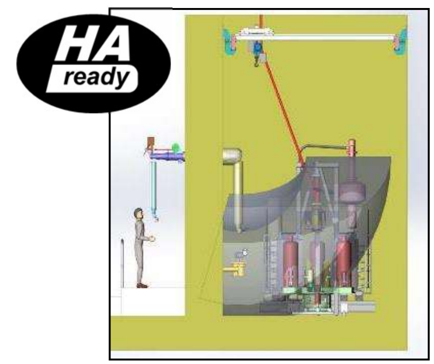
Smaller vitrification cells and very compact facilities



HLW facility
Two vitrification lines

Fully adapted for High Active waste

- Remotely operation & maintenance
- In-cell assembly and disassembly with moderate sized:
  - Overhead cranes
  - Master-slave manipulators
  - Remotely controlled tools



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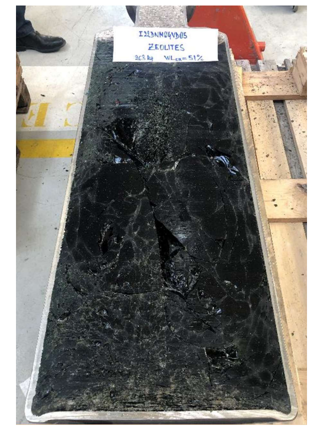
DEM&MELT main achievements

- **Several kinds of waste tested at different scale:**
  - Alpha liquid waste
  - Solid fission products deposits
  - Liquid fission products
  - Adsorbents (zeolite, silicotitanate, sand)
  - Ashes from incineration process
  - Sludges/Slurries from coprecipitation process
- **High Waste Loading achieved (up to 80% oxide)**
- **Very low radionuclides volatility**

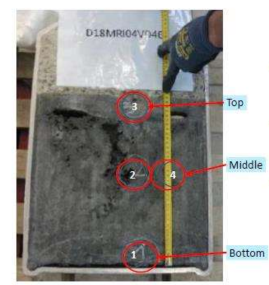
← Mix of zeolites, silicotitanates and sand
- WLox*: 60 wt.%
- Volatility: 0.04 % for Cs and 0.01 % for Sr

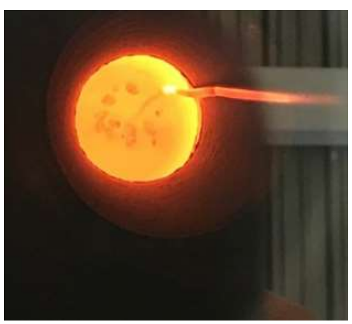


↑ Natural chabazite-type zeolites
- WLox*: ~ 50 wt.%
- Cs volatility: 0.08 wt%

↑ ALPS ferric and carbonated slurries
- WLox*: ~ 40 wt.%
- Sr volatility: 7.10-3 wt%

→ Mix of zeolites, slurries, silicotitanates and sand
- WLox*: 80 wt.%
- Volatility: <0.5 wt% for Cs and <0.1 wt% for Sr





$* \, WL_{ox}(\%) = \frac{\text{sum of oxide masses composing the waste (kg)}}{\text{mass of waste form (kg)}} \times 100$

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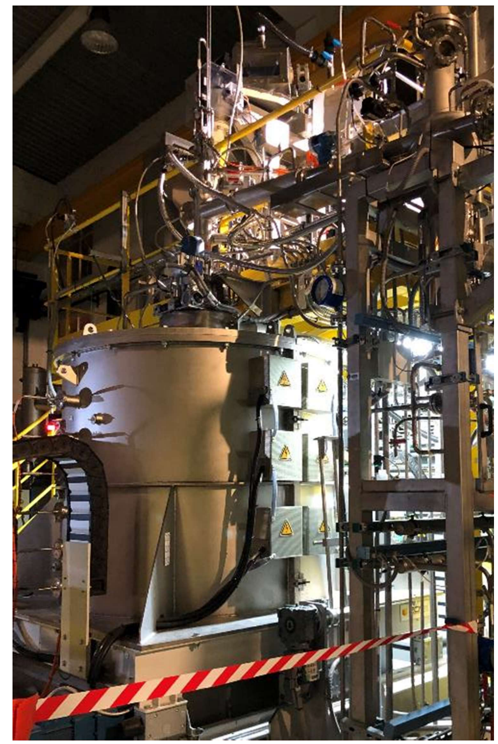
Vitrification of Fukushima Daiichi ALPS slurries

FULL SCALE PILOT TEST



Waste: ALPS ferric and carbonated slurries

- Highly **viscous** and **sticky** ➔ Very challenging regarding the feeding system
- High tendency to **foam** because of the high **gas release**
- High content of Sr
- Test 1 ➔ Programmed stop and go of the feeding system
- Test 2 ➔ Nominal operation with higher feeding rate

Waste surrogate made by coprecipitation and filtration, same as the real ALPS slurry

Waste surrogate in the feeding hopper

Full scale pilot

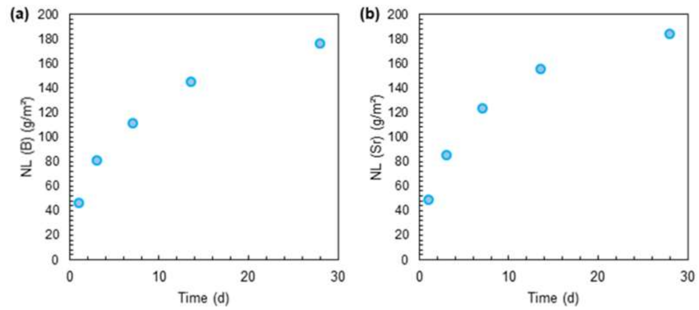
This test has been performed by the funds from the Japanese Ministry of Economy, Trade and Industry as The Subsidy Program "Project of Decommissioning and Contaminated Water Management"

Vitrification of Fukushima Daiichi ALPS slurries

FULL SCALE PILOT TEST

Waste: ALPS ferric and carbonated slurries

- Proper operation of the feeding system even during transient modes
- No foaming issue
- **WLox*: ~ 40 wt.%**
- **Sr volatility = 7 10^{-3} wt%**



Leaching test results

Test 1 **Test 2**

Half-cut canister

This test has been performed by the funds from the Japanese Ministry of Economy, Trade and Industry as The Subsidy Program "Project of Decommissioning and Contaminated Water Management"

$$* \boldsymbol{WL_{ox}}(\%) = \frac{sum\ of\ oxide\ masses\ composing\ the\ waste\ (kg)}{mass\ of\ wasteform\ (kg)} \times 100$$

Summary

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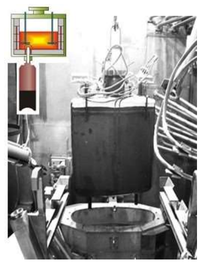
Summary

- **R7 and T7 outstanding records of operation & plants availability with respect to safety and glass quality**

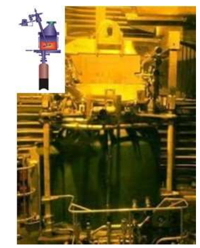
IHMM

- In operation since 1989 at LH plant
- ~ 24 100 canisters produced
- ~ 368 10^6 TBq vitrified
- ➚ Noble Metals incorporation
- ➚ Glass throughtput
- ➚ Melter (IHMM) lifetime



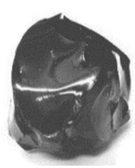
CCIM

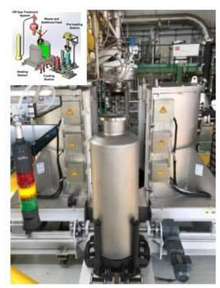
- In operation since 2010
- Retrofitted in a IHMM vitrification cell
- ~ 1 000 canisters produced
- ➚ Noble Metals incorporation & Glass throughtput
- Industrial maturity
- Since 2020 ➔ Vitrification of FP from commercial activity



- **Very High-Level Waste management with significant incorporation rate into the glass ➔ Around 1 Ci/g**

- **DEM&MELT In-Can Vitrification Technology**
  - Full-scale pilot commissioned in 2020
  - Benefiting from over 40 years of HLW vitrification
  - Simplicity, robustness, and versatility
  - High rates of waste incorporation into the matrix





Outcome of the continuous improvement policy implemented through a close collaboration between R&D, engineering teams and the industrial operator

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