Current technologies nuclear waste vitrification furnaces / Technology and issues

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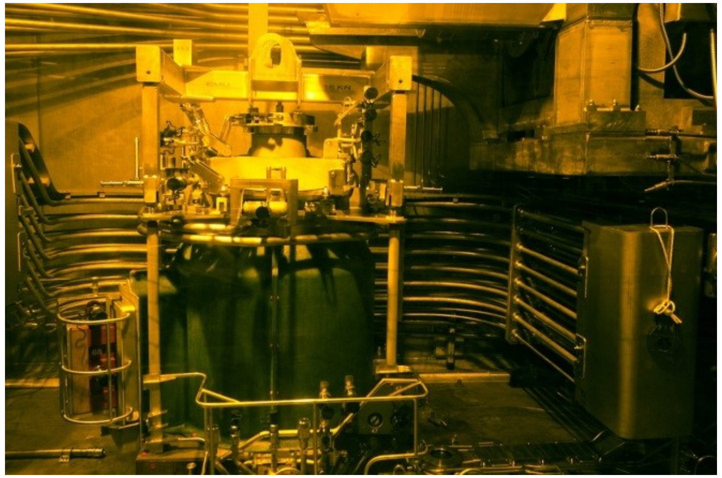
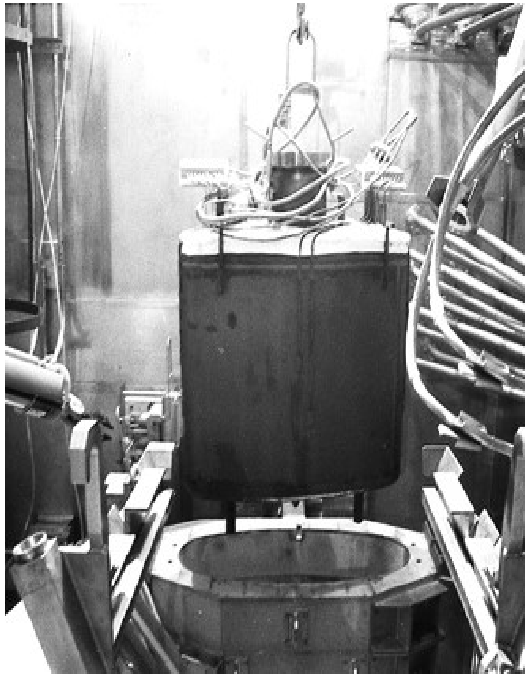
Summary

1. Introduction
2. French radioactive waste vitrification technology
3. Ceramic melter around the world
4. Difficulties arising with the Noble Metal Particles
5. Conclusion



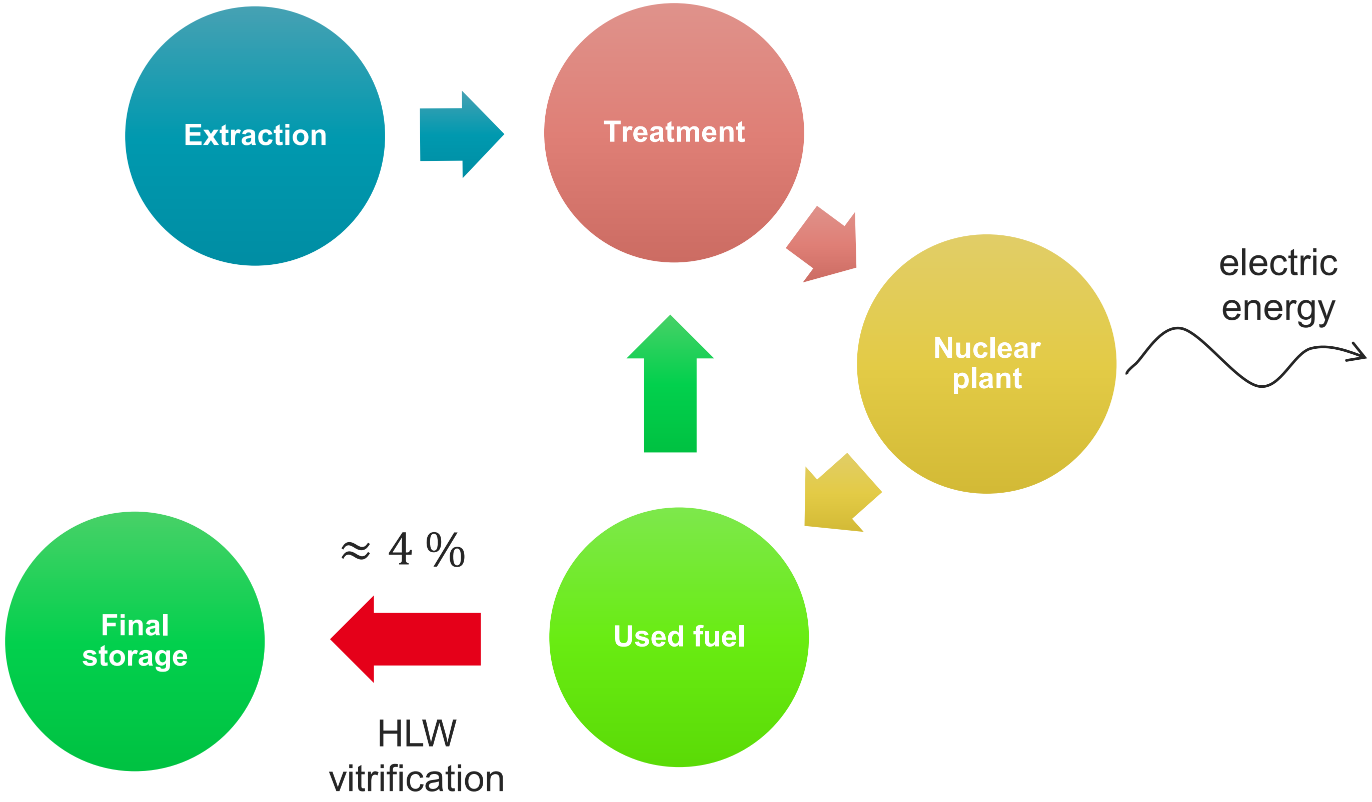
Cold Crucible Inductive Melter



Inductive Hot Metallic Melter

The vitrification process in the nuclear fuel cycle

- To ensure the long-term containment of High-Level and Long-Lived Radioactive Waste (HLW), primarily consisting of:
  - Fission Products
  - Minor Actinides



Glass canister

Example of chemical composition of a waste solution to be vitrified (UOx type)

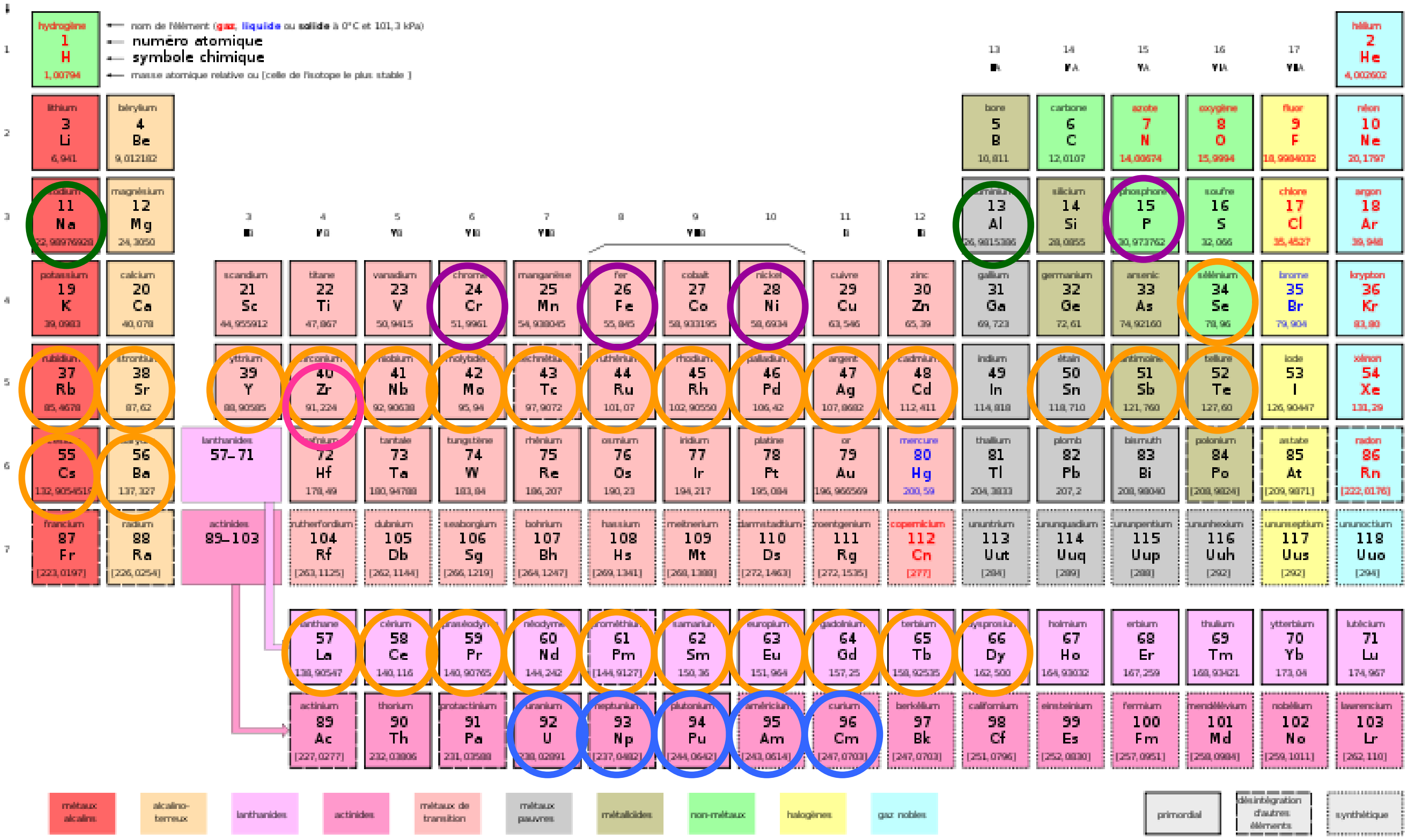
Fission product

Actinides

Corrosion and degradation products

Adjuvants

Fines de cisaillage



Which choice of confinement matrix?

- **Organic or cement-encased materials:** excluded due to activity level

- **Mineral materials**

  ✓ **Crystalline materials: initial research focus (late 1950s). Difficulty in incorporating all Fission Products**

  ✓ **Glassy materials (from the 1960s).**

    - Well-known material, used for centuries
    - Archaeological analogues available
    - Stability under irradiation
    - Chemical durability, good corrosion resistance
    - Physico-chemical properties in line with the incorporation of a broad chemical spectrum of radionuclides
    - Industrial feasibility

Vitrification processes - Definition

- Functions to be ensured:

1. Water removal: Evaporation, drying (100°C)
2. Transformation of elements into oxides (300°C to 800°C)
3. Introducing constituents of the glass matrix
4. Glass production: Reaction between materials and fusion (1050°C to 1250°C)
5. Gas treatment
6. Glass package production and storage

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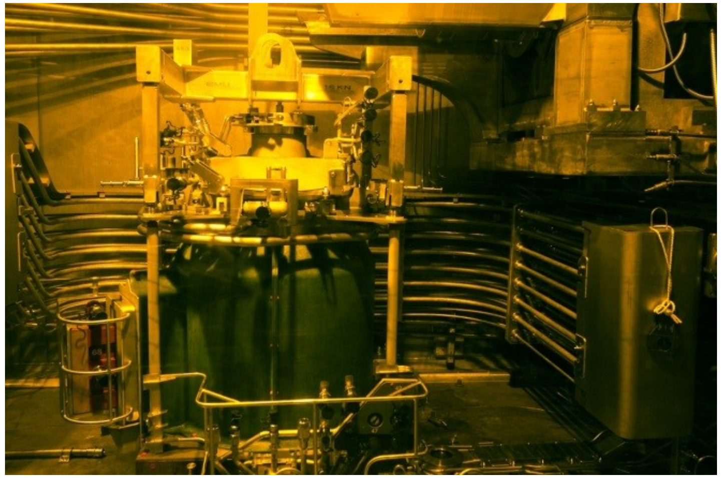

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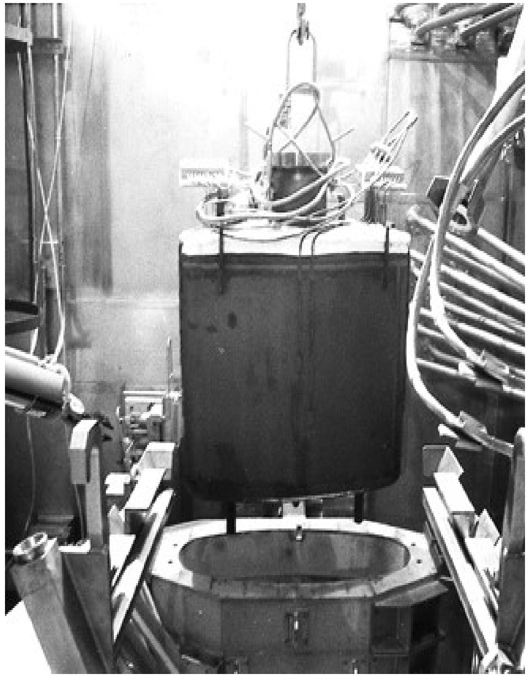
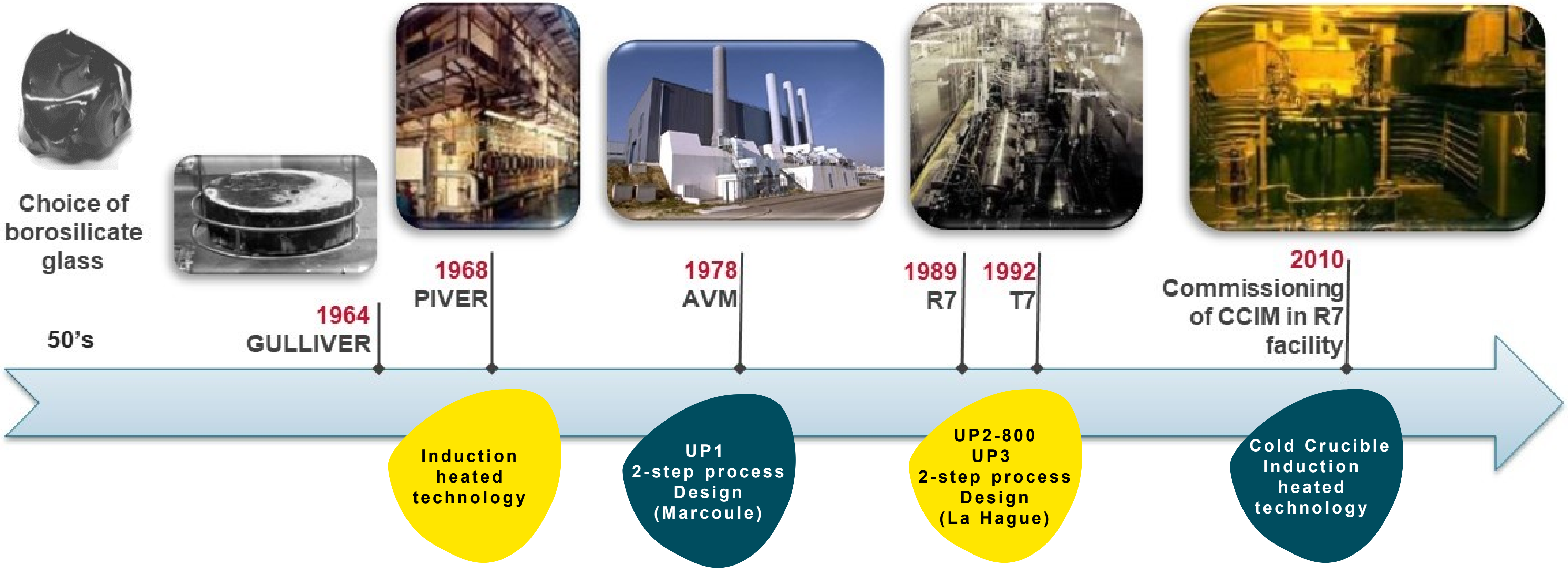


Inductive Hot Metallic Melter

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



Orano's Vitrification Industrial Experience

Over 40 years of Industrial Operation

AVM Marcoule – Operated by Orano from 1978 to 2009

1 vitrification line

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

R7 / T7 La Hague Plant – Operated by Orano since 1989

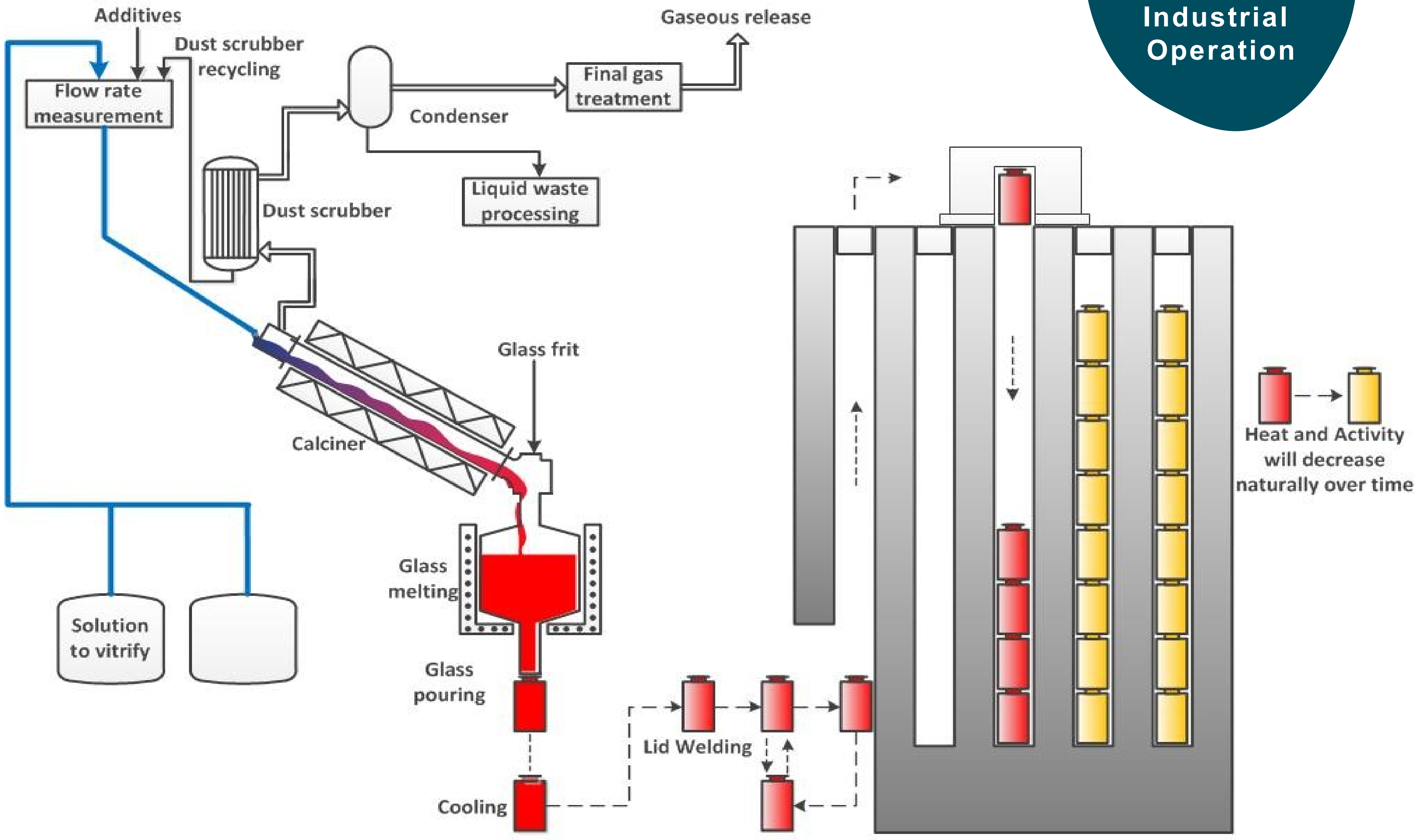
2 vitrification facilities (6 lines)

IHMM (end of 2022)

- ~ 24 700 canisters produced
- ~ 9 900 metric tons of glass produced
- ~ 377 10^6 TBq vitrified

CCIM (end of 2022)

- ~ 1 000 canisters produced



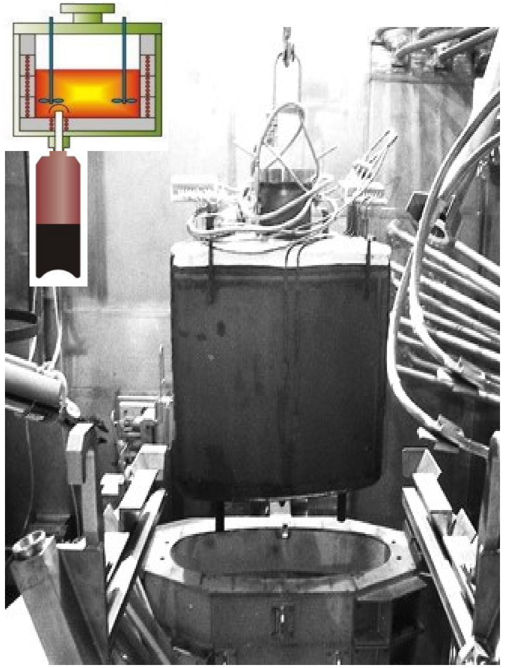
Main Vitrification Technologies Developed in France

CCIM

Cold Crucible
Direct induction

In operation since 2010

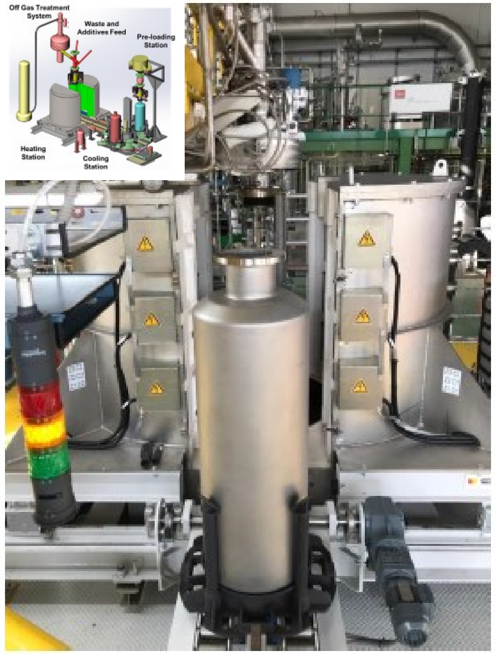
IHMM



Metallic Melter
Indirect induction

In operation since 1978

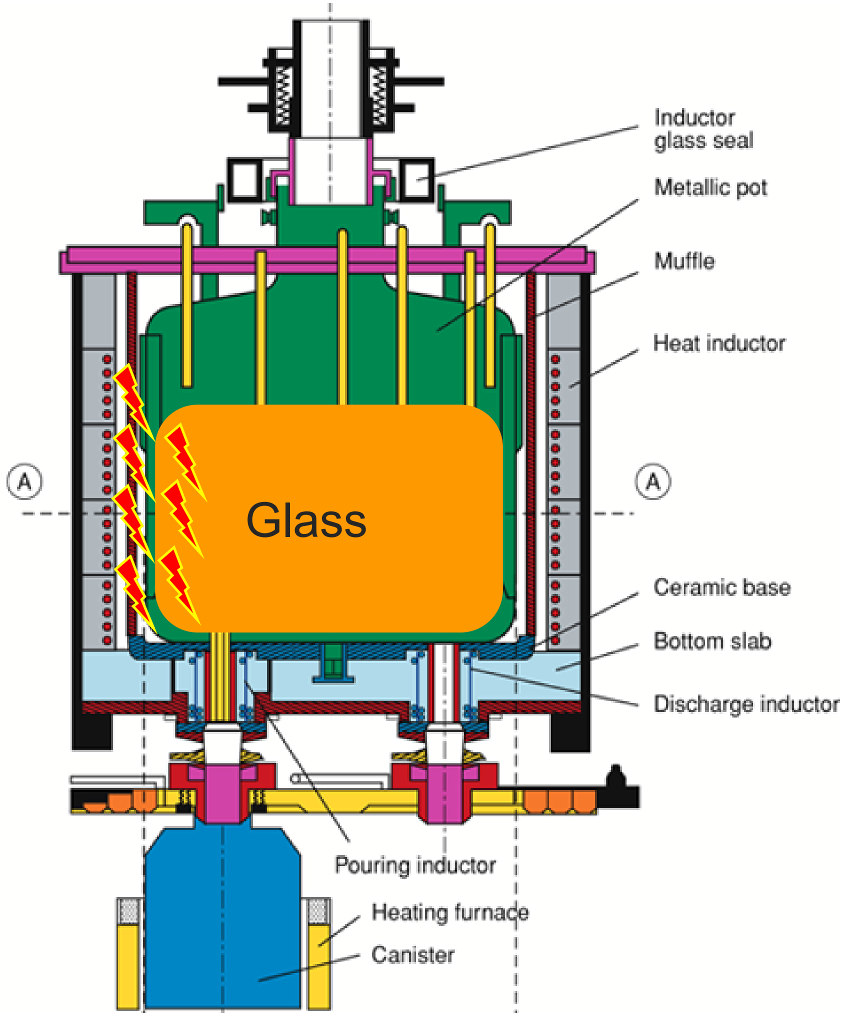
In-Can Melter



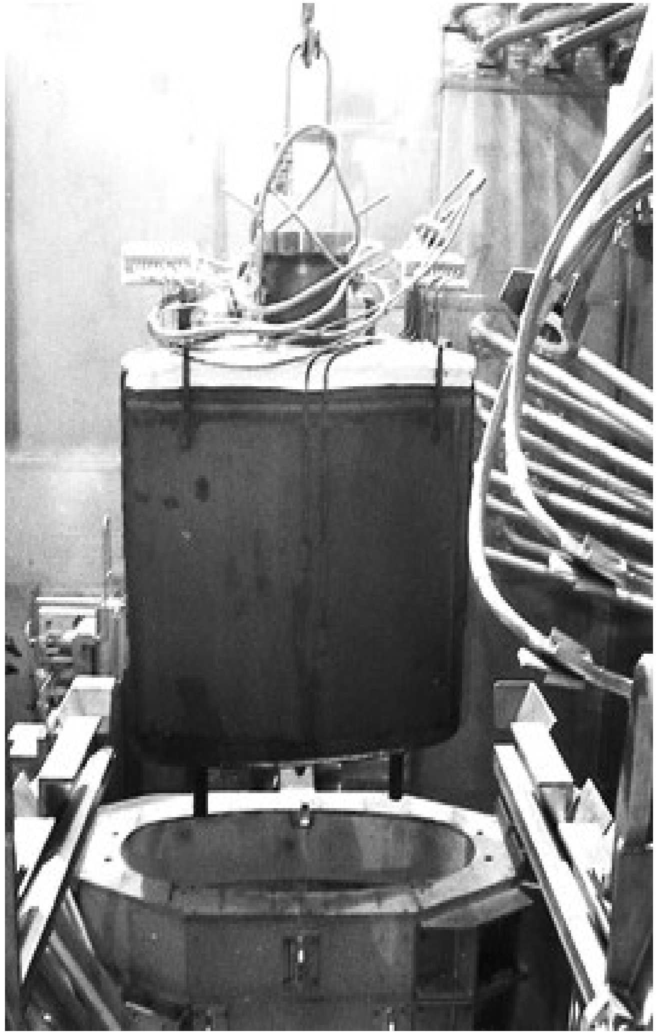
Resistance heating
Thermal homogenization

Full scale pilot commissioned in 2020

Induction Heated Metallic Melter



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

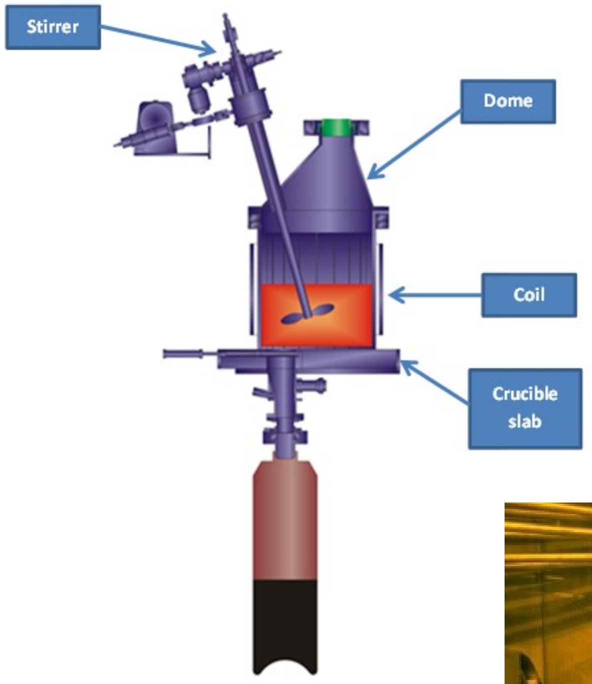
TRL 9

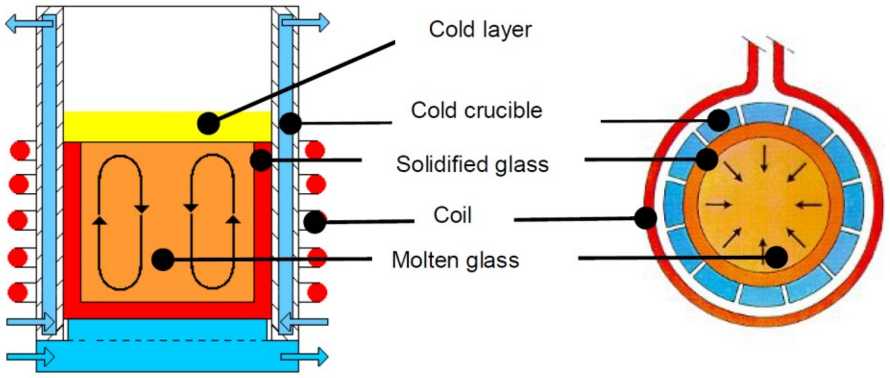
- Over 40 years of industrial operation
- Over 11 100 metric tons of glass produced

Wasteform

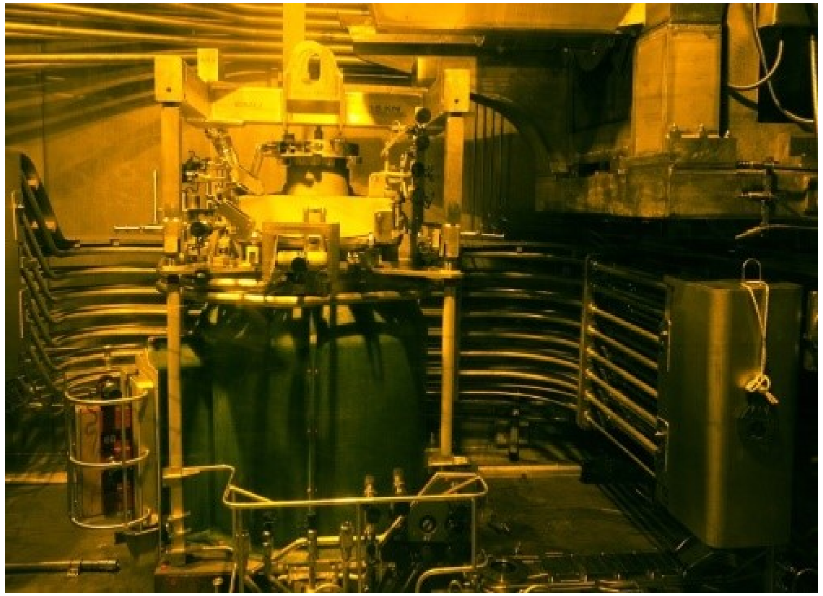
- Homogeneous borosilicate glass
- Around 1 Ci/g

Cold Crucible Induction Melter





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°)

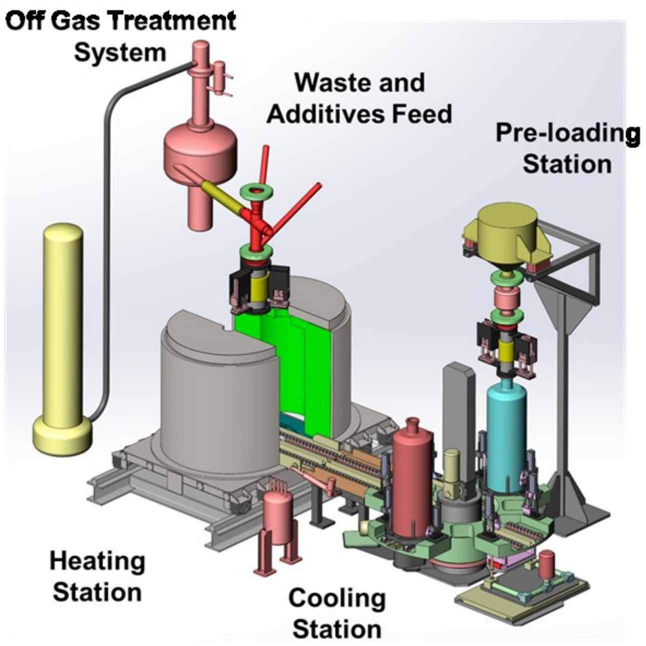
TRL 9

- Over 14 years of industrial operation

Wasteform

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

DEM&MELT In-Can Melter



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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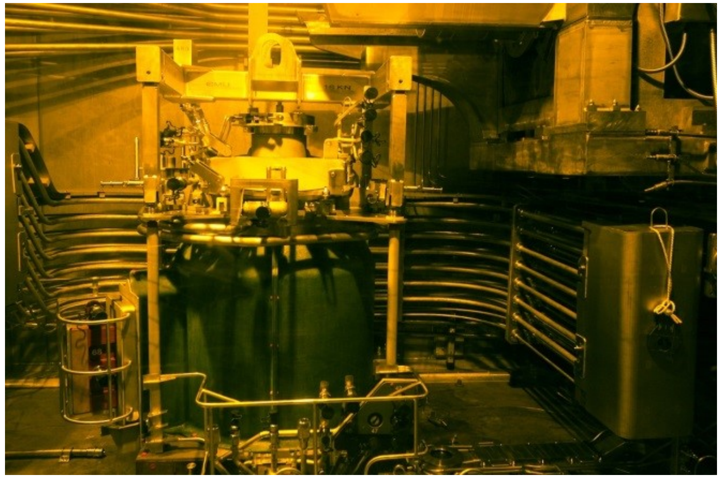

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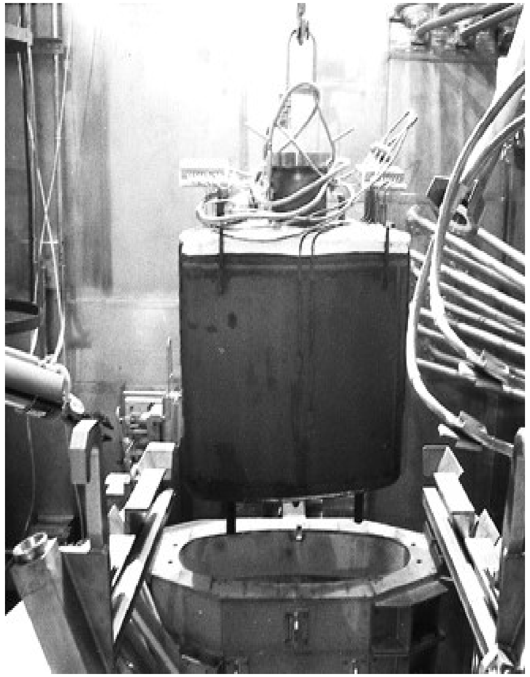
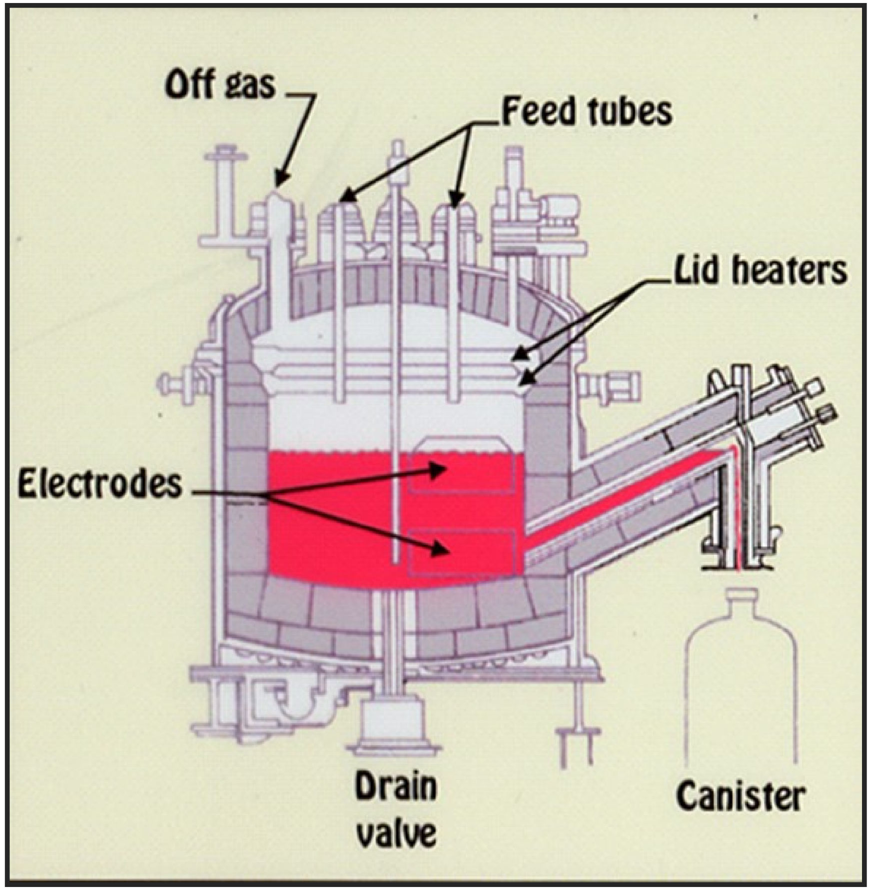


Inductive Hot Metallic Melter

USA : Defense Waste Processing Facility (Savannah River – USA)

Atelier DWPF

- First melter from 1996 to 2002 replaced by a second improved melter in operation during 12 years.
- Third melter in operation since 2017
- Empty mass 65 t
- Waste to be treated: 130,000 m3 of sludge in 51 tanks of military origin
- Different glass formulation for each tank
- Very few Noble Metal particles
- Capacity from 70 to 100 kg/h
- Installation of bubbler rods to improve productivity
- 4,200 canisters have been poured (8000 tons of glass)

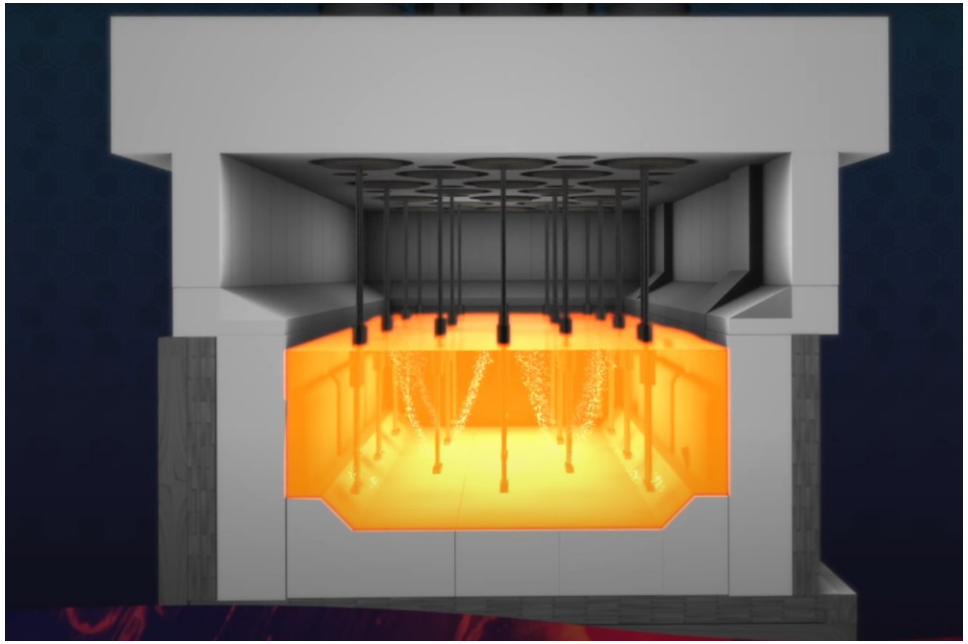


https://www.energy.gov/em/articles/defense-waste-processing-facility-reaches-25-years-successful-operations-srs

USA : Hanford VIT plant

- **Two plants : Low level Waste facility and High level Waste facility**
- **LLW facility**
  - **300 tons furnace**
  - **18 bubblers rods**
  - **Flat bottom**
  - **No Noble Metals particles**

https://melterheatup.hanfordvitplant.com/



https://youtu.be/NOcpthpN3g0

The LLW facilty is in the starting operations
The HLW facility is under construction and will produce an annual average of 480 canisters.

Russia : Mayak EP-500/1R ceramic melter

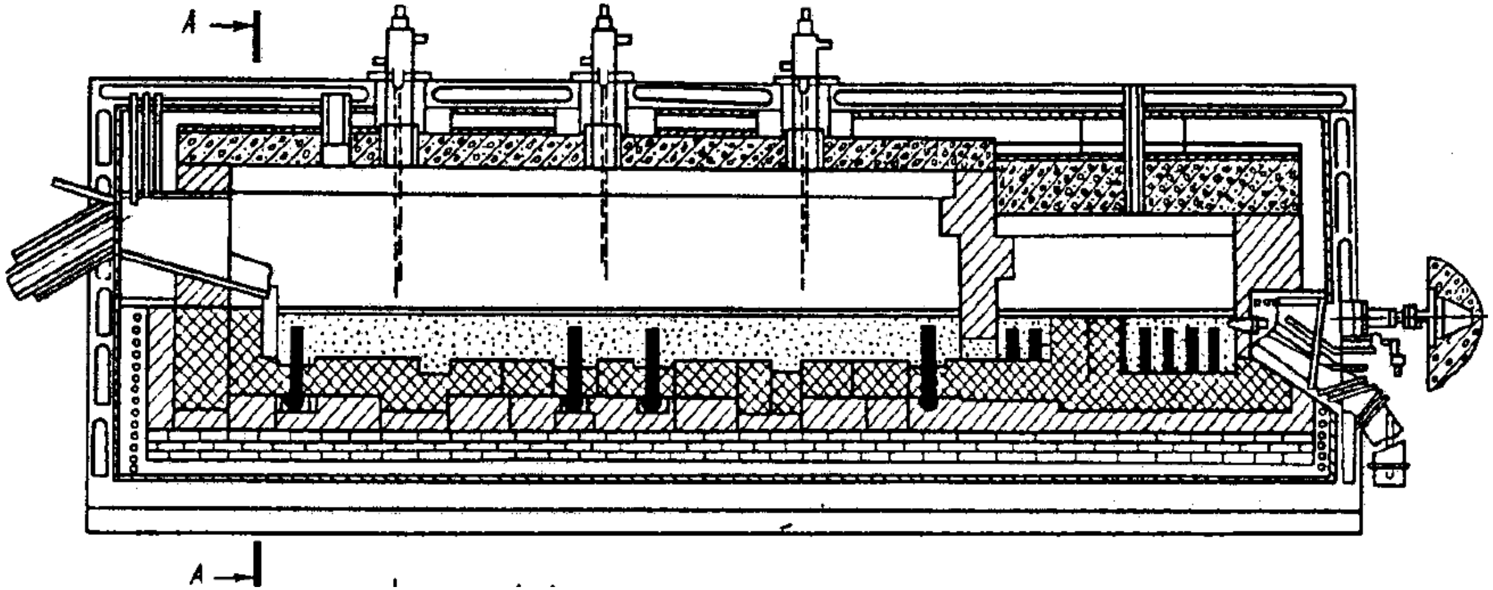
Vitrification since 1987

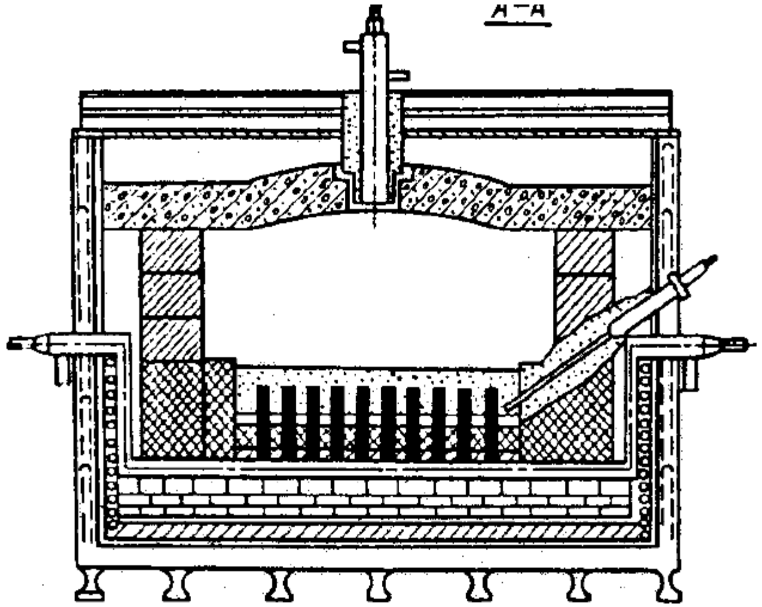
Four succesive ceramic melters

Molybdenum electrodes in the bottom

More than 4000 tons of glass produced

Low content of NM <0,1 %w

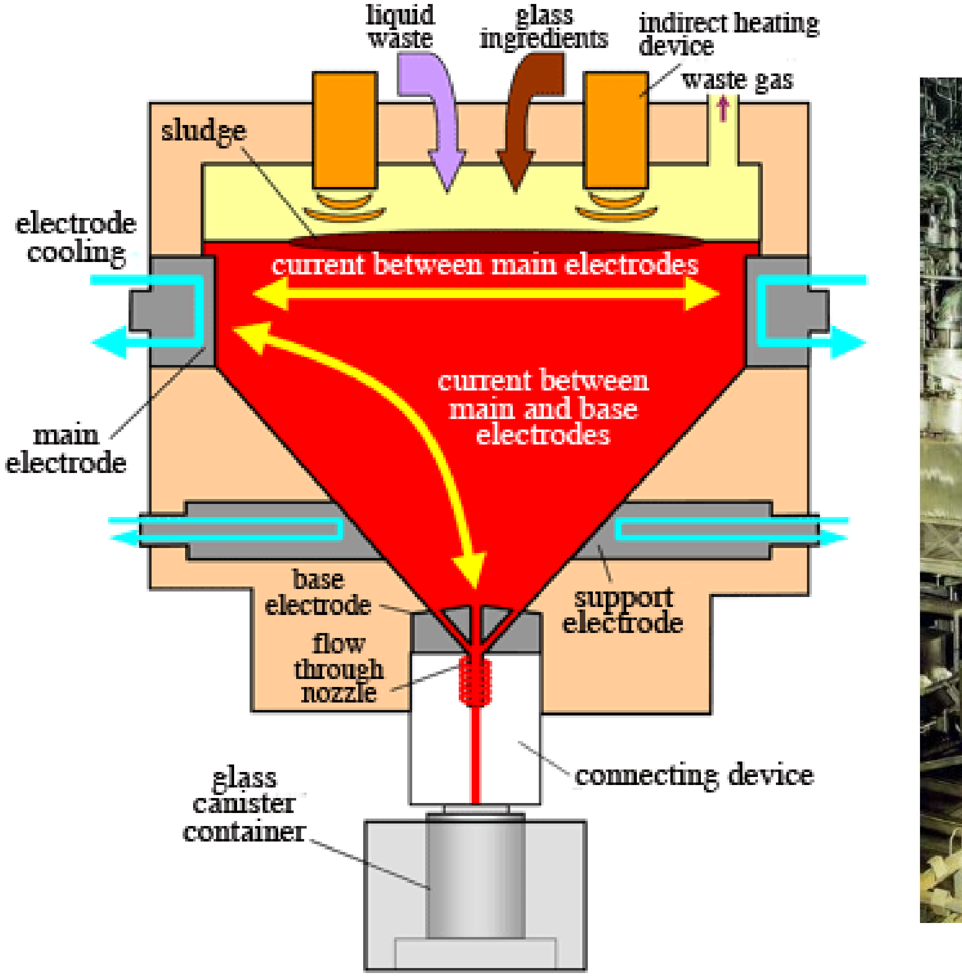




Japan : Tokai Vitrification Facility

TVF – (Tokaï mura)

- Tokai plant reprocessing solutic (light water reactors)
- Two successive furnaces
  - 1995 to 2002
  - 2004 to 2007
- Production 8.5 kg/h but reduced 6.5 to manage Noble Metals particles
- Empty mass : 15 tons
- 100 tons of glass produced
- Bottom slope 45°

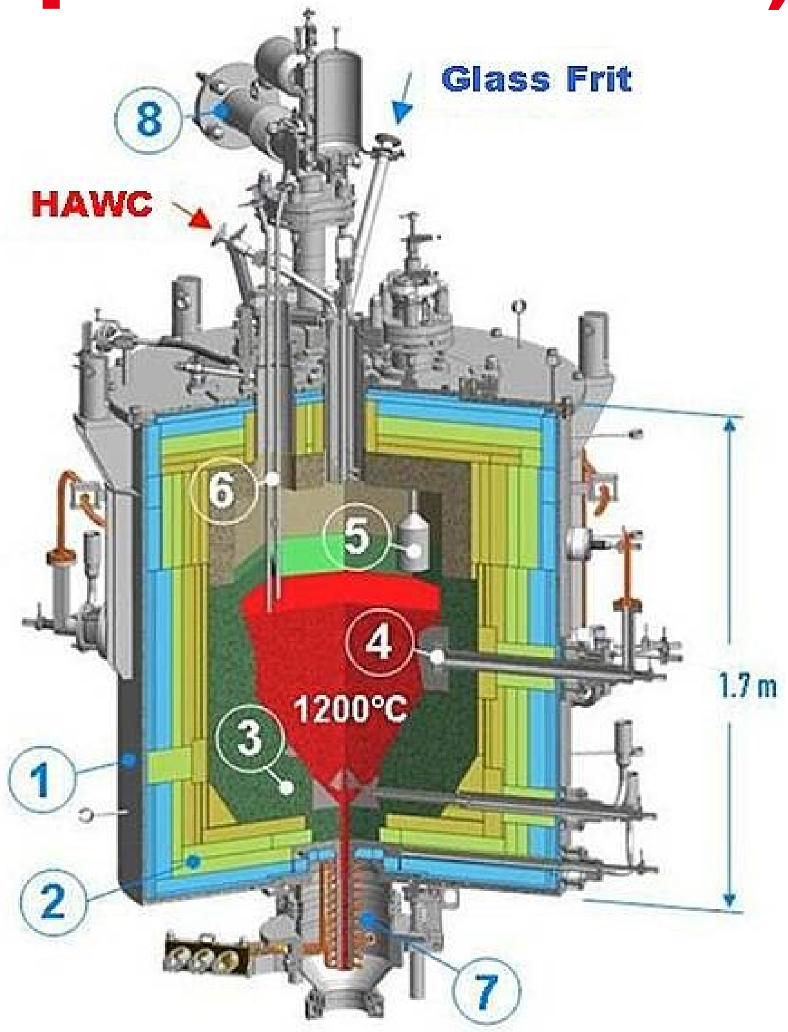


High Active Liquid Waste Vitrification Equipment Outline (Glass Melting Furnace)

Germany : VEK (WAK plant Karlsruhe)

Features

- 2008 - 2009
- Steeply inclined bottom
- 9 month of operation
- Production of 5 to 7 kg/h
- 50 tons of glass produced
- NM <1%
- Bottom slope of 65°
- Accumulation of NM after 7 months
- Installation of bubbling
- Glass rinse without Pts



Major characteristics of the melter	
Outer diameter	1.5 m
Height	1.75 m
Weight	9 Mg
Nominal throughput	10 l/h
Glass production rate	7 kg/h
Melting pool volume	400 kg
Glass per batch	100 kg
Duration of a filling campaign	1.5 h
Filling frequency	every 15 h

http://www.ewn-gmbh.de/ewngruppe/wak/decommissioning-projects/karlsruhe-reprocessing-plant-wak-plant/vitrification.html?L=1

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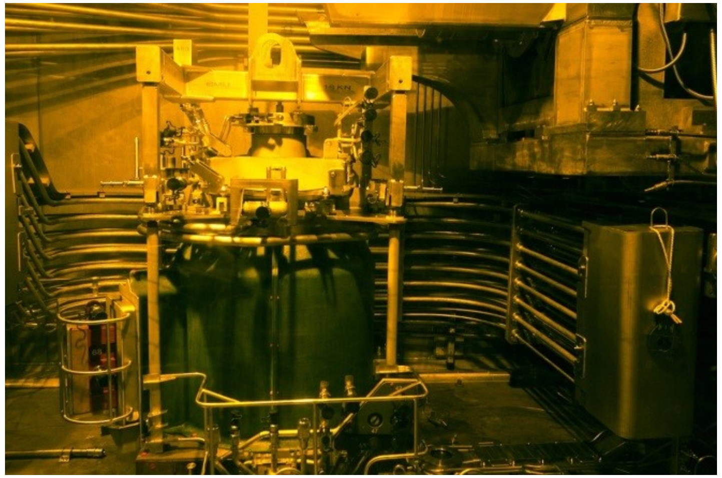

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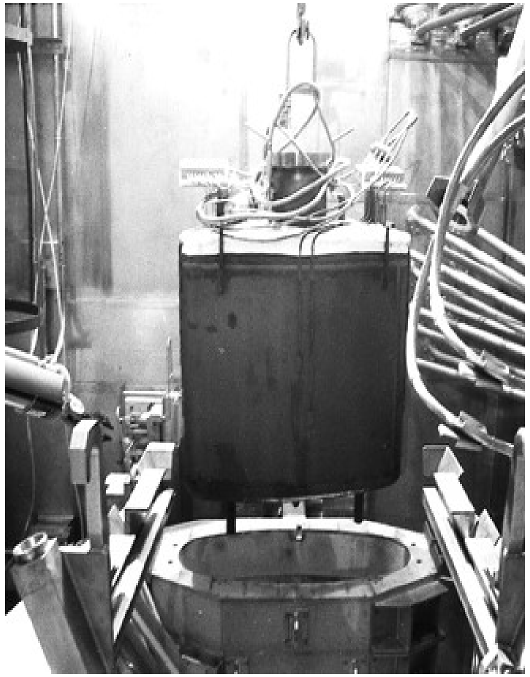
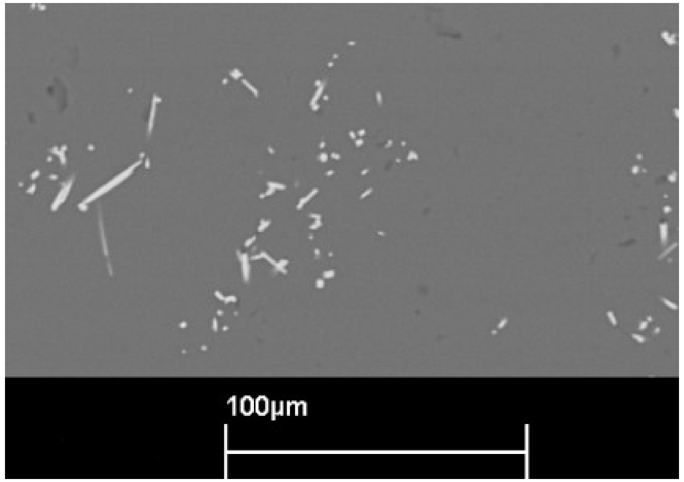


Inductive Hot Metallic Melter

Noble metal particles

SEM picture of inactive nuclear glass



Platinum Group Metal particle (PGM)

- In the nuclear glass, noble metal particles are present
  - Concentration depending on the waste type (0.1 to 3 %w)
  - Ruthenium dioxide (RuO_2) needles
  - Palladium Rhodium ($Pd\ Rh$) spheres

- Strong impact on the vitrification process via the physical properties of the glass :

i. **Viscosity** → impact on the mixing quality of the glass

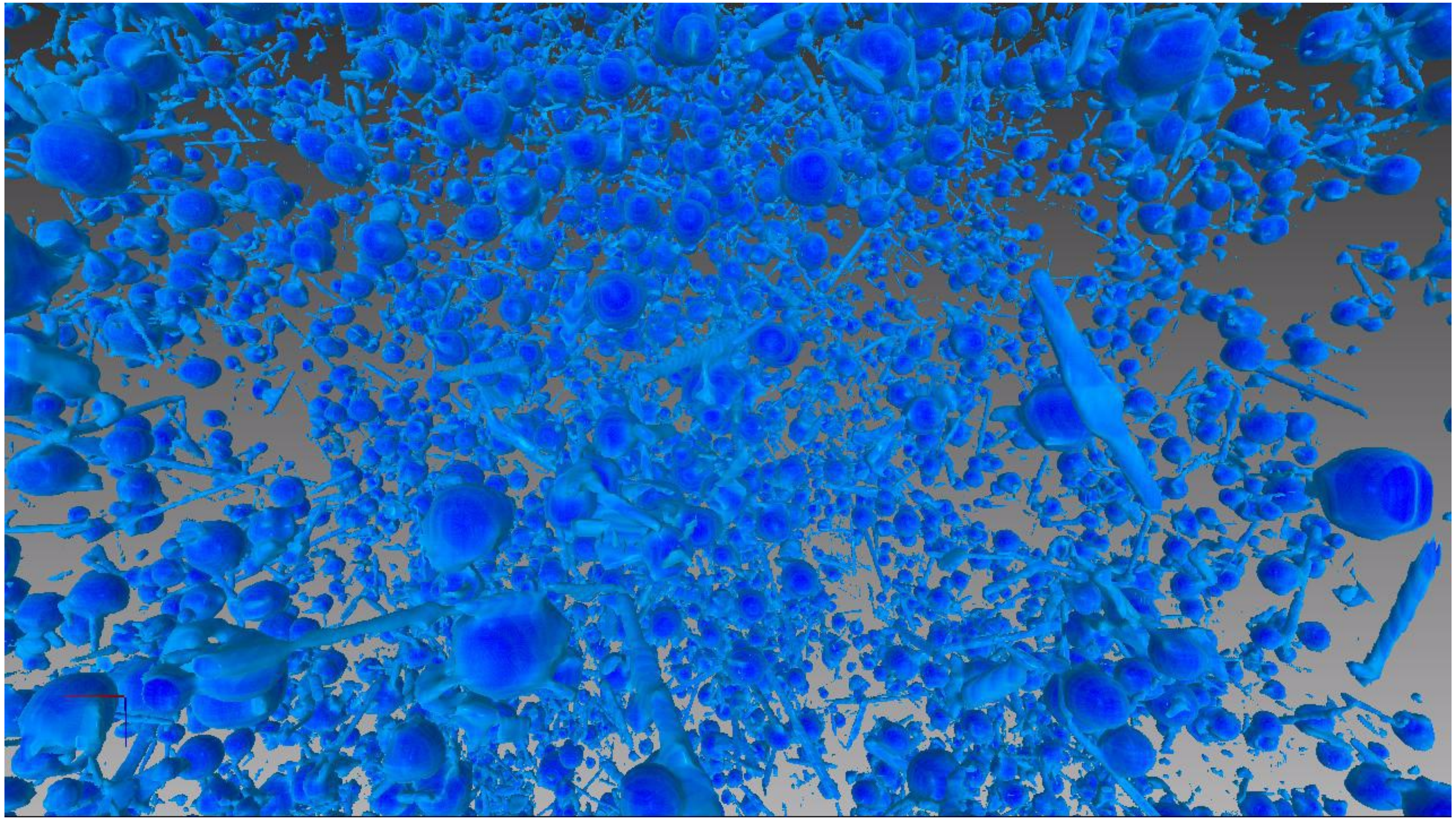
ii. **Density** → impact on the sedimentation / settling risk

iii. **Electrical conductivity** → impact on the induction heating

Noble metal particles

X Micro-tomography

- Samples of inactive glass send to the European Synchrotron Radiation Facility (ESRF, ID19)
- Voxel size of $0{,}16^3\ \mu m^3$



Description

Needles : Ruthenium dioxide (RuO_2)

Spheres : Palladium (Pd)

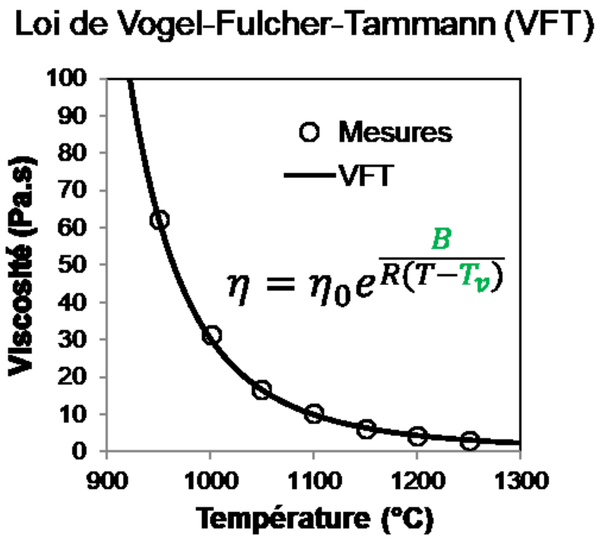
- Diameter of one sphere : ~ 5 µm

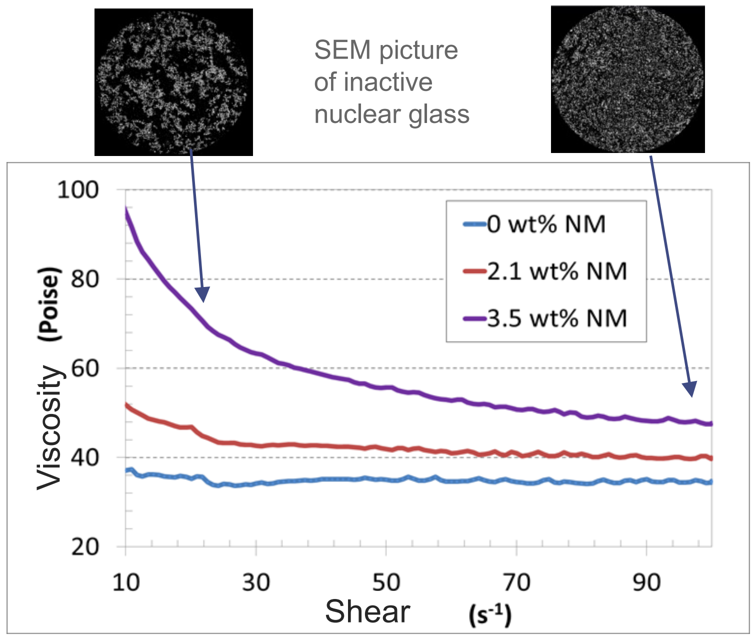
Volume fraction of particules in this sample : 3,5 %v (~10%w PGM)

Noble metal particles

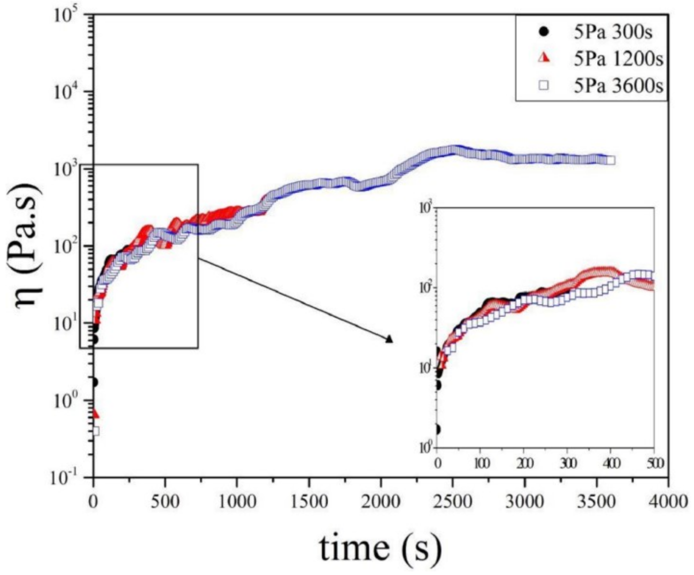
Noble Metal particles effect on the viscosity

- The temperature dependence of the glass viscosity is well modeled with an VFT law
- A non Newtonian behavior of the viscosity is observed with NM particles
- Particles tend to aggregate under low shear conditions leading to an increase in apparent viscosity
- In fact, the structuration of the particles is quite a slow phenomenon (several hundred seconds)





Caroline Hanotin, *et al.* (**2016**), **Journal of Nuclear Materials**, 477.



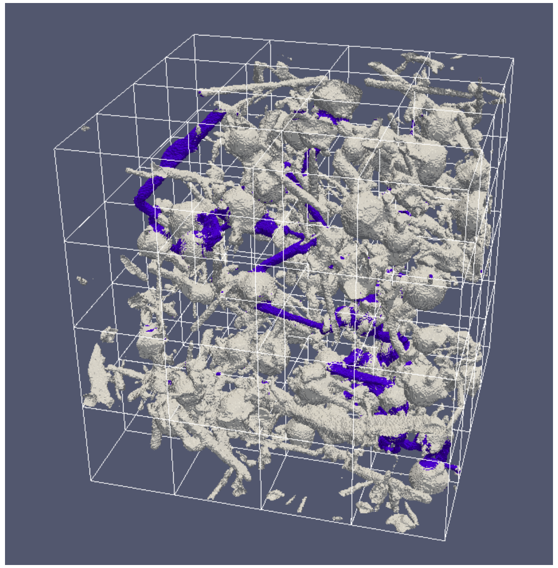
Machado, Norma Maria *et al.* (**2022**). **Journal of Nuclear Materials**. 563.

Noble metal particles

Noble metal particle effect on electrical conductivity of the nuclear glass

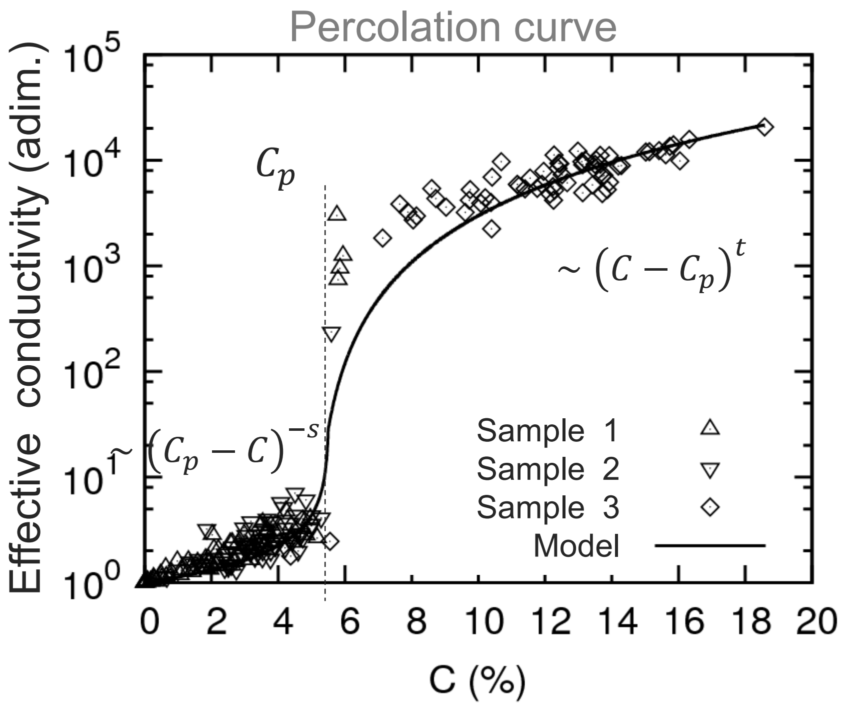
- The law of effective electrical conductivity shows a percolation threshold concentration : Cp

Example electrical circulation in the Noble Metals particles (gray)



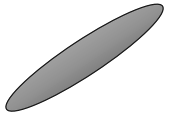
Barba Rossa G, Grenoble University, PhD 2018

Percolation curve



Garboczi *et al.* 1995

$$C_p \approx \frac{0.6}{\Psi}$$



aspect ratio $\Psi \approx 10$

$$\sigma = f(C, C_p, s, t)$$

Japan : KA vitrification plant (Rokkasho Mura)

- Vitrification of the Rokkasho reprocessing nuclear used fuel plant (800t CU/year)
- 40 tons empty
- Bottom slope at 45°
- <u>Noble Metal particles content at ~1 w%</u>
- Around 4000 kg of molten glass
- Theoretical production of 43 kg/h
- Maximum production reach : 28 kg/h
- Significant difficulties in managing NM which create short circuits between electrodes

- The “plant acceptance” of the vitrification furnaces has still not been finalized. The factory is at a standstill.

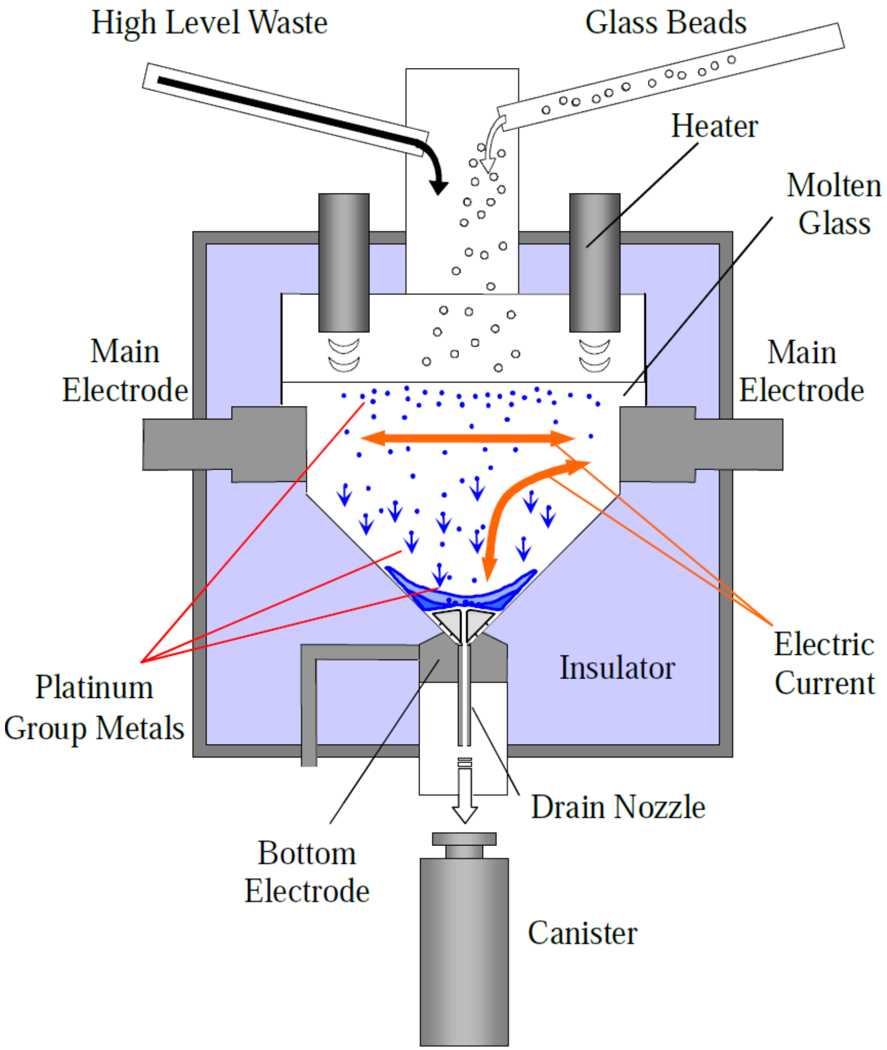
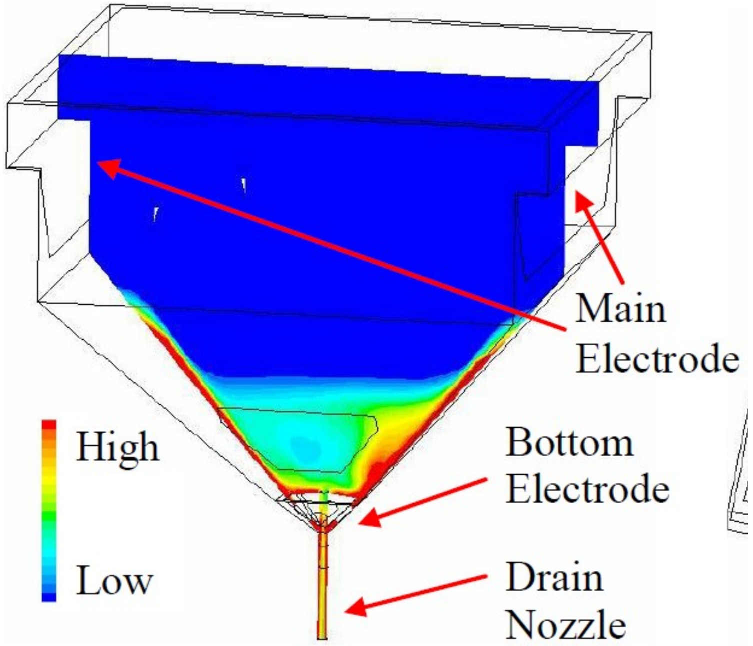


Figure 1. HLW Glass Melter

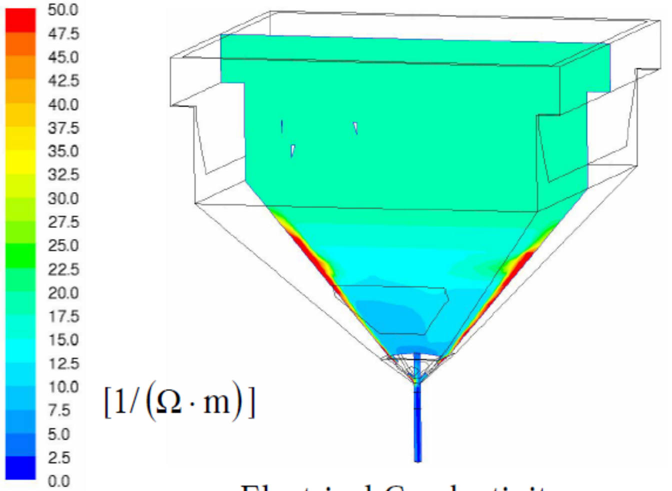
Yoshiyuki, ISO; MATSUNO, S.; UCHIDA, H.; Isamu, OONO; FUKUI, T. & Takaaki, OOBA
Proceedings of the International Conference on Nuclear Engineering, **2007**, doi=10.1299/jsmeicone.2007.15._ICONE1510_116

Japan : KA vitrification plant (Rokkasho Mura)

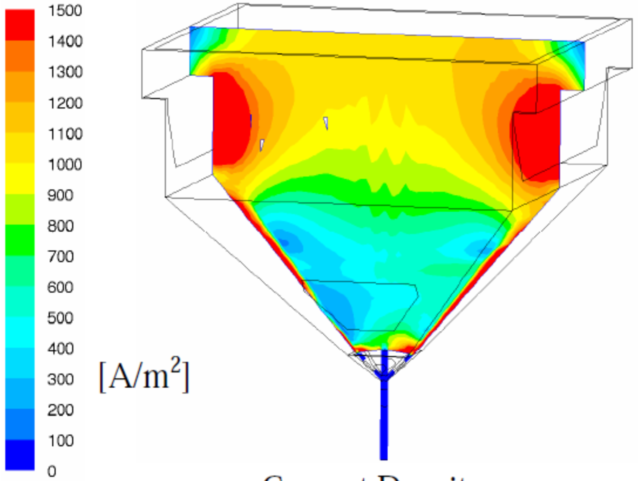
- Modelisation of the Noble Metals particles in the melter
  - Sedimentation of NM
  - Enhancement of local electrical conductivity of the glass
  - Distortion of electrical current path and Joule effect efficiency
  - Risk of short circuit or overheating and degradation of the refractory



PGM concentration



Electrical Conductivity



Current Density

Yoshiyuki, ISO; MATSUNO, S.; UCHIDA, H.; Isamu, OONO; FUKUI, T. & Takaaki, OOBA *Proceedings of the International Conference on Nuclear Engineering,* **2007**, doi=10.1299/jsmeicone.2007.15._ICONE1510_116

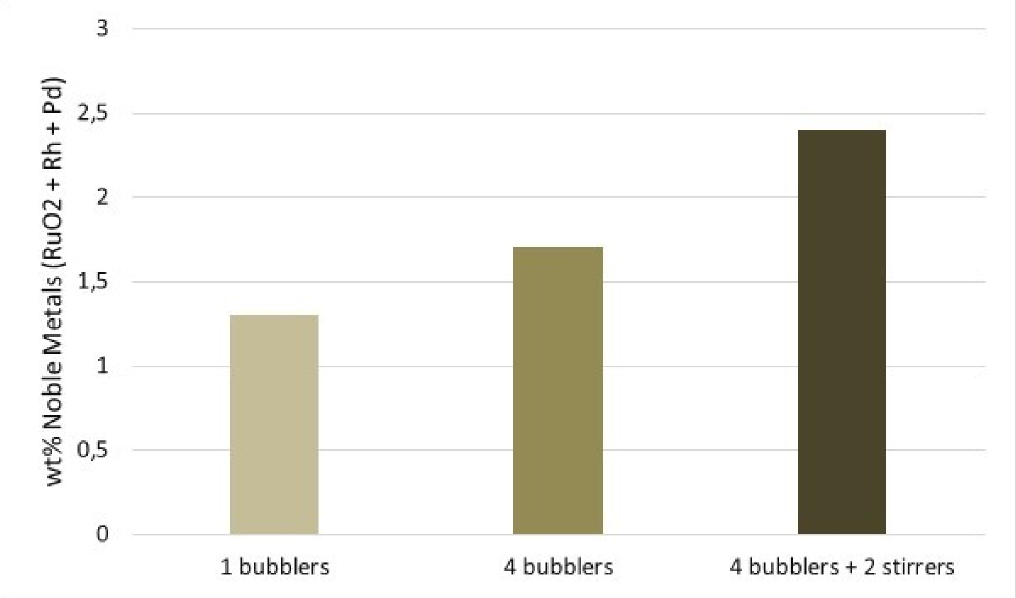
France : Noble Metals Incorporation Achievements IHMM

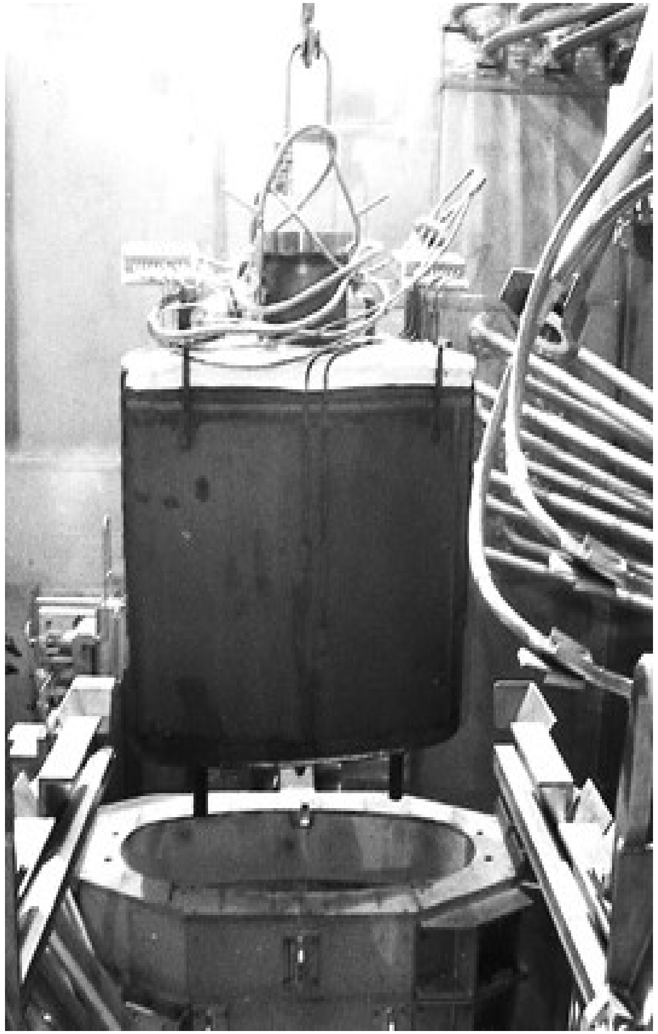
Several technological enhancements implemented to the melter design to improve mixing capability

- 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

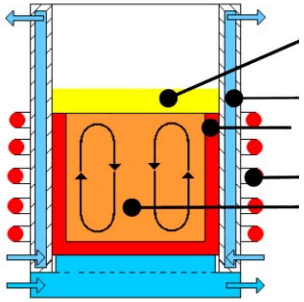
Operating parameters optimizations to increase NM incorporation as well as glass throughput

- 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





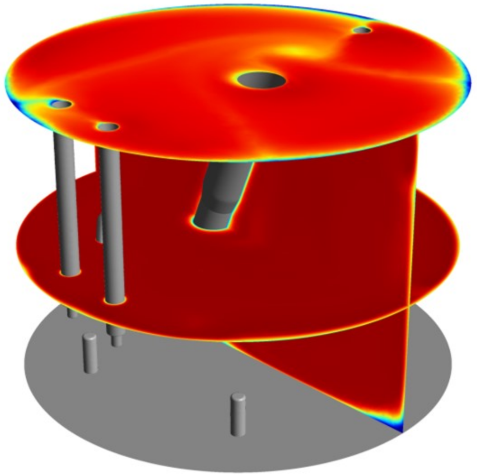
France : Noble Metals Incorporation Achievements CCIM



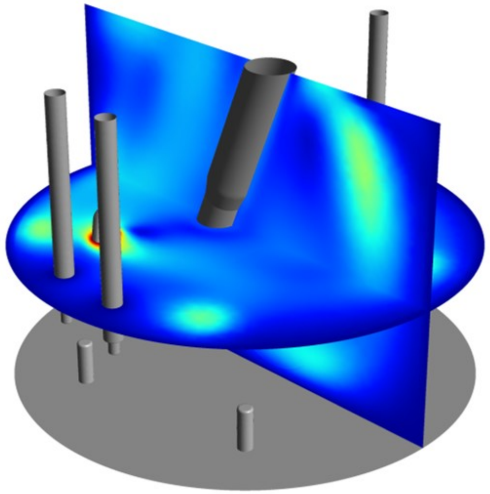
Design of mixing in the Cold Crucible melter has take into account the NM particles problem from the beginning

- Mechanical stirring and gas bubbling promote a good thermal homogeneïty and prevent any NM sedimentation
- UOx glass with up to 2.6 wt% of NM (RuO_2, Rh, Pd) is produced at La Hague plant with CCIM
- Numerical simulation has helped to understand and design the mixing in the CCIM

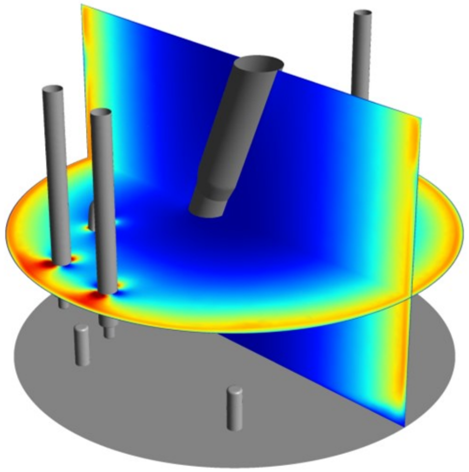
Temperature, velocity and Joule power density fields in the glass



$T \in [1200; 1480]$ K

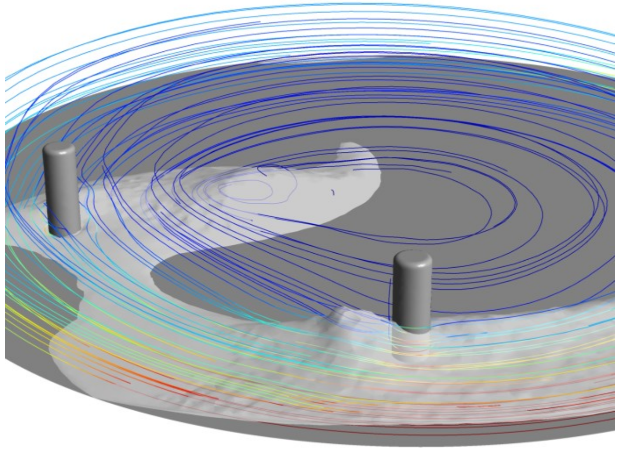


$U \in [0; 1]$ m/s

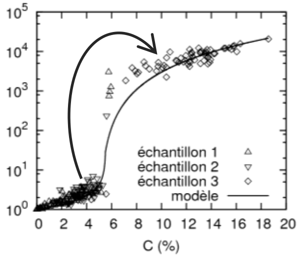


$p_J \in [0; 4]$ MW/m^3

Iso-contour of NM concentration and Induced electric current in the glass



Iso-contour $C = 5{,}3$ %v



Conclusion

- All glass melters for nuclear waste vitrification are full electric due to off-gas treatment
- Ceramic melter with different electrode configuration are used in several country in the world.
  - They are suitable for nuclear glass composition without Noble Metals particles
- In France, induction melter are used (indirect and direct) because of their compactness
  - Induction heated metallic melter has proven its ability to produced HA nuclear glass with outstanding records of operation & plants availability and with respect to safety and glass quality
  - Cold crucible induction melter is efficient to produce highly corrosive glass and with a high content of NM

IHMM

- In operation since 1989
- ~ 24 700 canisters produced
- ~ 377 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
- High throughput
- Treatment of highly corrosive glass melts
- High NM incorporation

CCIM is also suitable
to produce non nuclear glass
of very high purity !
We perform melting tests
at the CEA Marcoule

Thank you for your attention

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