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Joint ICTP-IAEA Workshop on Fundamentals of Vitrification and Vitreous Materials for Nuclear Waste Immobilization

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ICTP

IAEA

# NUMERICAL SIMULATIONS IN THE DEVELOPMENT OF THE FRENCH RADIOACTIVE WASTE VITRIFICATION PROCESSES USING INDUCTION FURNACE

PRESENTATION BY EMILIEN SAUVAGE

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# Summary

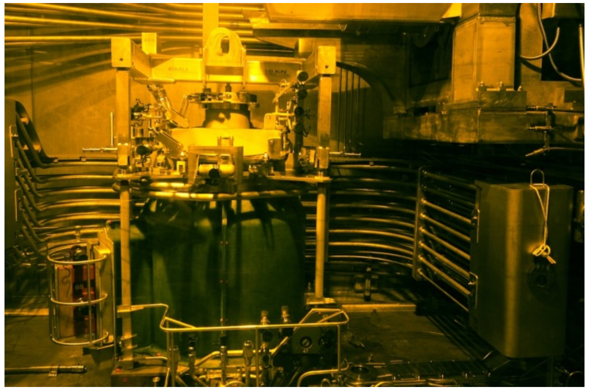
1 − French radioactive waste vitrification technology presentation

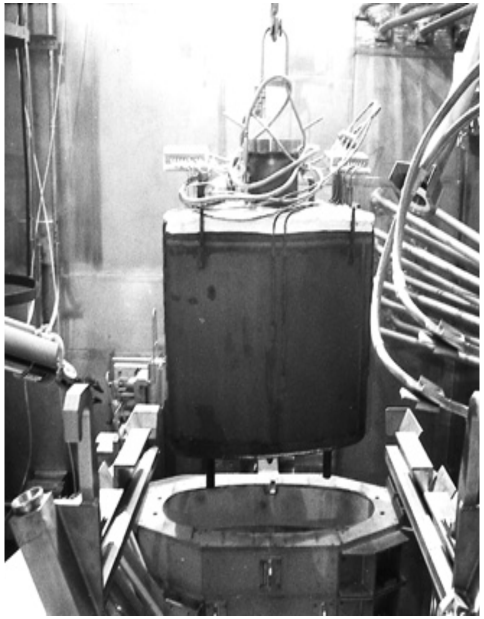
2 - Vitrification processes simulation

- 2-a Induction heating modeling
- 2-b CFD modeling
- 2-c Coupled physics
- 2-d Noble metal particles

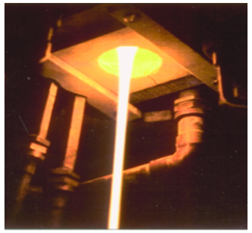
3 – Ongoing development

4 - Conclusion





# CONTEXT

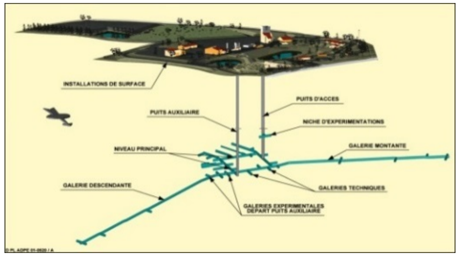


- **France ➔ high-level liquid waste arising from nuclear fuel reprocessing successfully vitrified for more than 30 years**
  - ➲ Durable containment of the long-lived fission products
  - ➲ Minimization of the final waste volume
  - ➲ Operational performance achieved in vitrification plants

- **Areva La Hague plants start in 1989-1992**
  - ✓ **Around 20 000 canisters produced (400 kg of nuclear glass each)**

- **Joint Vitrification Laboratory (LCV) ➔ CEA** (French Alternative Energies and Atomic Energy Commission) and **AREVA** Common research laboratory in charge of qualifying new processes and matrices for waste containment

- Need of **higher throughputs** and **higher temperature : in 2004 AREVA NC decision to implement a CCIM in R7**



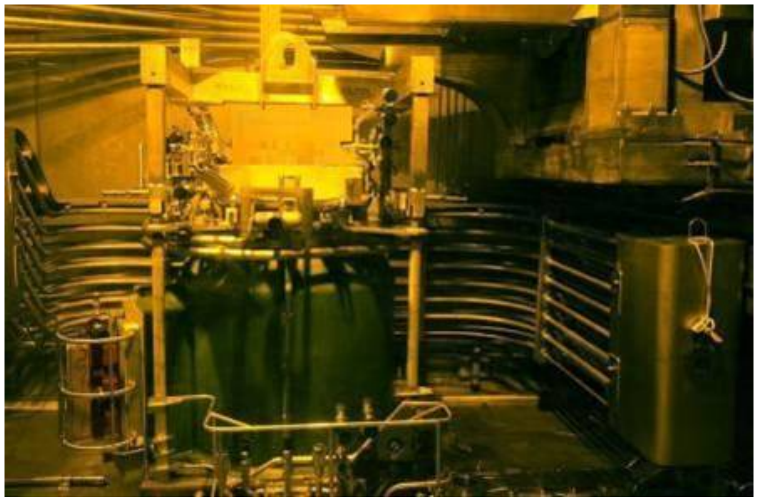

➔ Design and qualification of the **Cold Crucible Induction Melter (CCIM)** technology by the CEA Marcoule

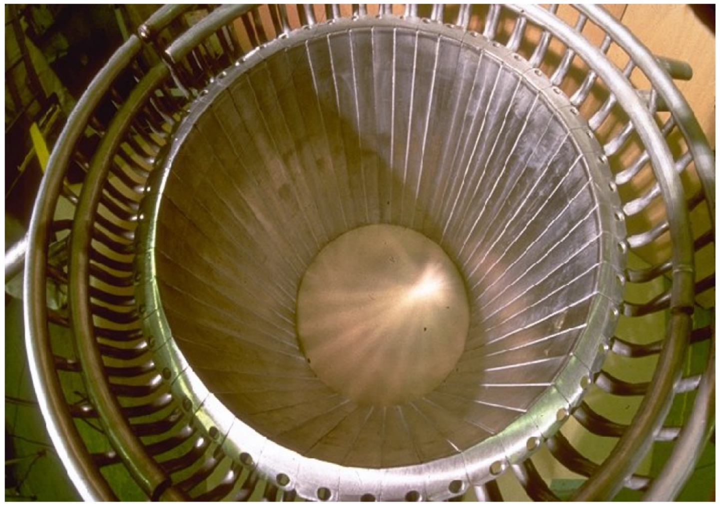
# CONTEXT

- AREVA has replaced one existing Induction Heated Metal Melter (IHMM) in a production line in the R7 facility at La Hague plant by a Cold Crucible Induction Melter (CCIM)

- April 2010 ➔ Hot operation for the first time ever at the La Hague R7 vitrification facility

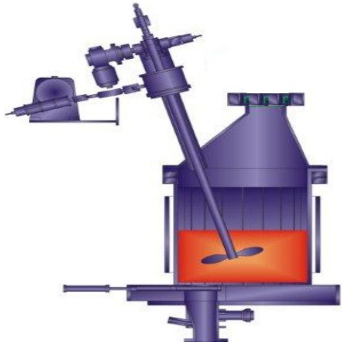
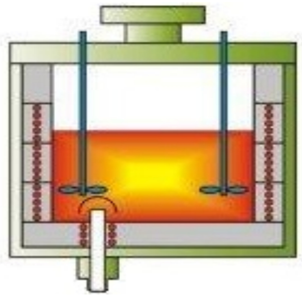
- CCIM in commercial operation for more than seven years, processing active effluents from D&D operations and high-level liquid waste from reprocessed U-Mo-Sn-Al spent fuel (GCR fuel)





# VITRIFICATION PROCESS OPERATED IN THE LA HAGUE PLANT

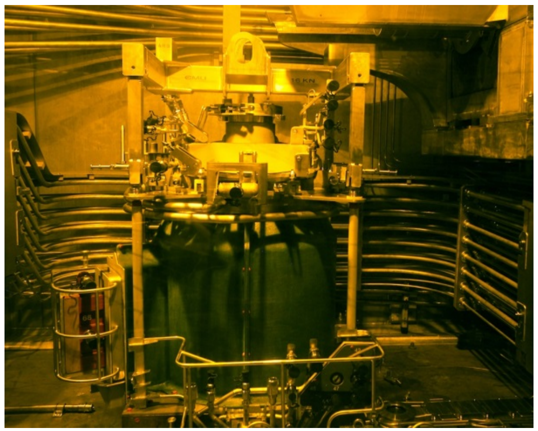
## Furnaces technology overview

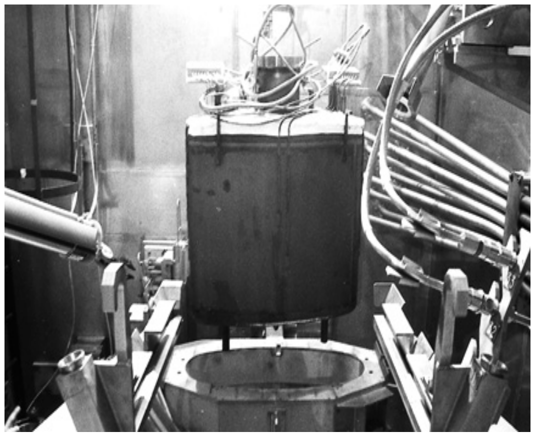
| | Cold Crucible Inductive Melter | Inductive Hot Metallic Melter |
|---|---|---|
| |  |  |
| **Characteristics:** | | |
| - Induction heating type | direct | indirect |
| - Number in LH plant | 1 | 5 |
| - Commissioning in | 2010 | 1989 |

Average chemical compositions for R7T7 glass produced in the industrial facilities at La Hague

| Oxides | Average composition of industrial glass (w %) |
|---|---|
| SiO2 | 45,6 |
| B2O3 | 14,1 |
| Al2O3 | 4,7 |
| Na2O | 9,9 |
| CaO | 4 |
| Fe2O3 | 1,1 |
| NiO | 0,1 |
| Cr2O3 | 0,1 |
| P2O5 | 0,2 |
| Li2O | 2 |
| ZnO | 2,5 |
| Oxides (PF+Zr+actinides) + Suspension of fines Actinide oxides | 17 |
| SiO2+B2O3+Al2O3 | 64,4 |

Picture in hot cell
(Areva La Hague)

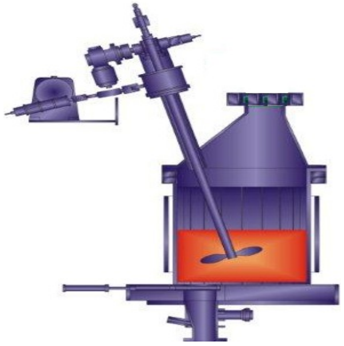


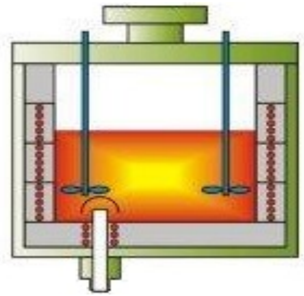


# VITRIFICATION PROCESS OPERATED IN THE LA HAGUE PLANT

## Furnaces technology overview

| Characteristics: | Cold Crucible Inductive Melter | Inductive Hot Metallic Melter |
|---|---|---|
| - Molten glass mass | 400 kg | 270 kg |
| - Max glass throughput | 45 kg/h | 25 kg/h |
| - Max temperature | > 1400°C | 1100°C |
| - Mechanical stirring | yes | yes |
| - Gas bubbling | yes | yes |
| - Approx. life time | > 2 years | 0.5 year |
| - Generator power | 600 kW | 400 kW |
| - Induction frequency | 300 kHz | 4 kHz |





IHMM

Inside view in CCIM (inactive Marcoule)

# Summary

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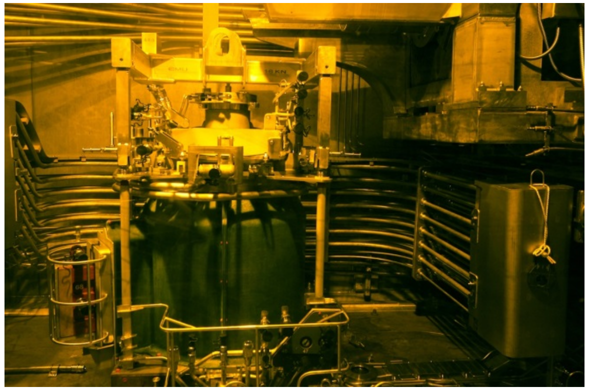
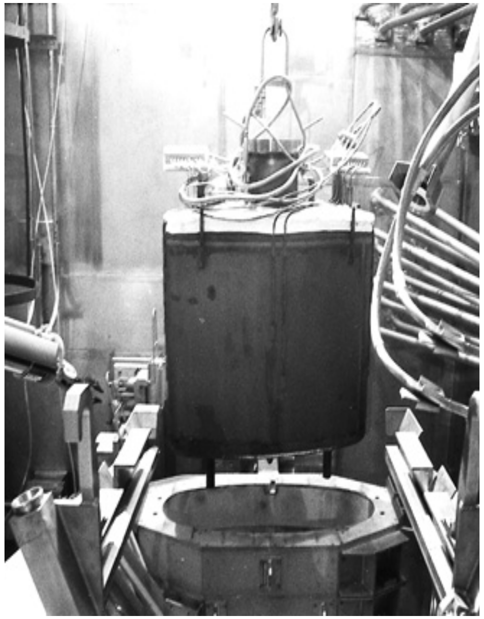




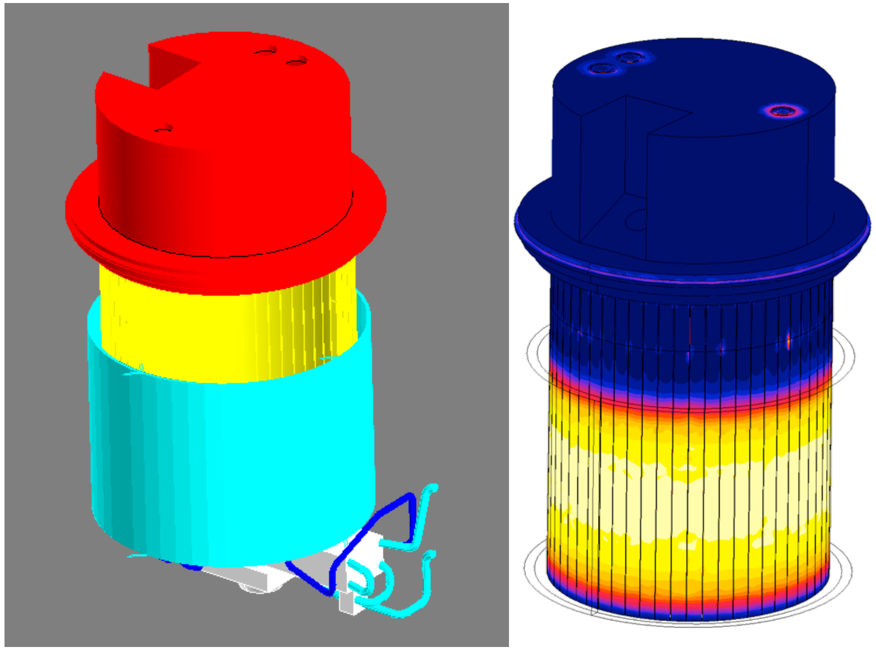
## Simulation of induction heating in vitrification processes

– Software : Flux® Induction equation
– Problems arise from the industrial scale and complexity of the process
– Validation of predictability of the electromagnetic simulation

Validation

| | | Unit | Expt. | Sim. |
|---|---|---|---|---|
| Inductor current | | A | 1981 | 1981 |
| Joule Losses | side | kW | 104 | 105 |
| | bottom | kW | 5.2 | 5.5 |
| Equivalent resistance | | mOhm | 27.8 | 28.1 |

Cold crucible | Magnetic field

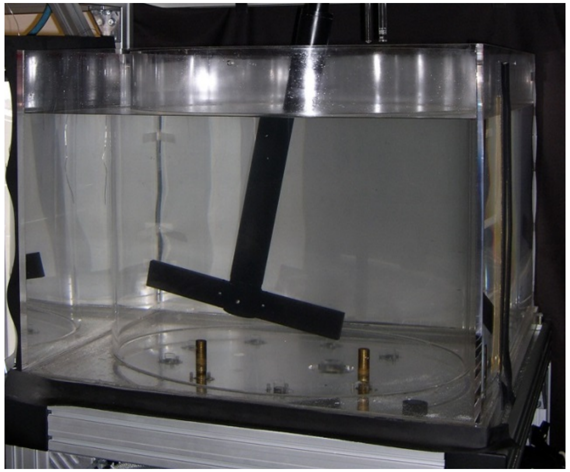


Main Use :
- Limitation of electrical external loop
- Design evolution to increase robustness and reliability

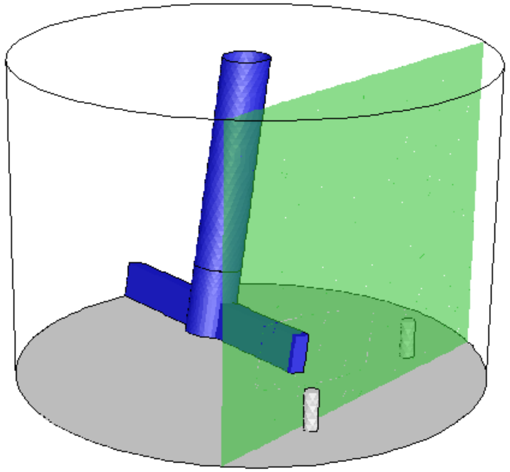
## CFD of the vitrification processes : mechanical stirring

- First CFD were axysimetric. Their interest was limited by the fact that mechanical strirring and gas bubbling were not taken into account.
  - Main development based on similiratity experiment in oil (scale 1)
  - Use of Particle Image Velocimetry (PIV) to measure oil velocity
- Fuent software. Sliding mesh technique give best results

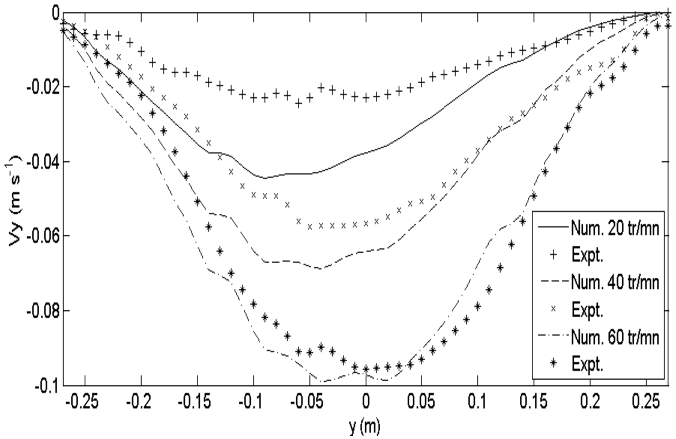
Similaririty experiment



3D CFD computing (sliding mesh)



Velocity profile for different rotating speed




Main Use :

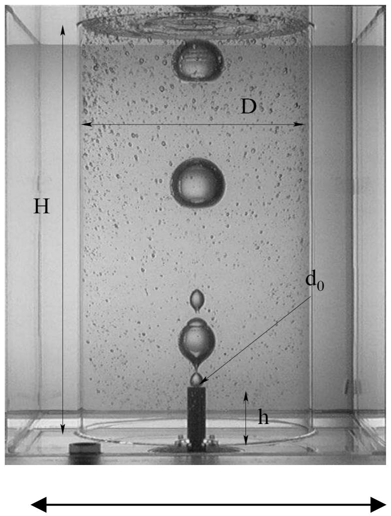
- Shape and position of stirrer, optimised rotating speed

## CFD of the vitrification processes : gas bubbling

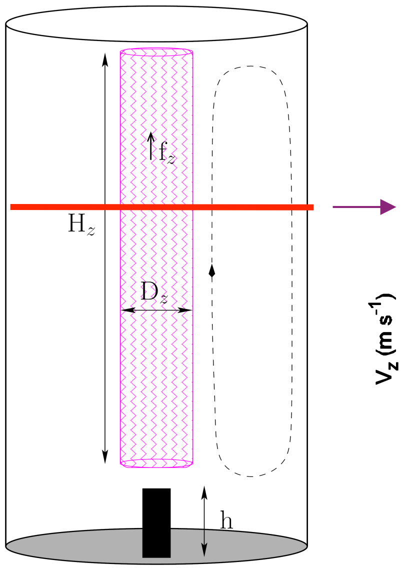
- Semi empirical modelling have been built
  - Based on correlation giving bubble diameter and velocity as a function of the airflow, fluid viscosity

Experiment in hydraulic similarity with silicon oil

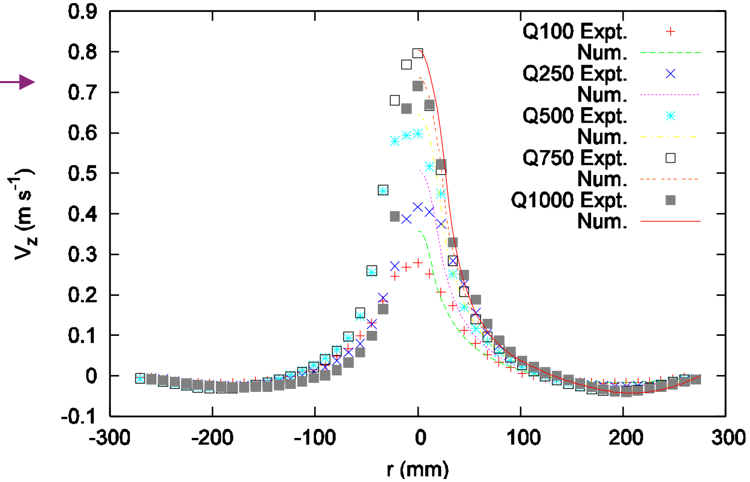
**R. RIVA *(CEA / Grenoble)***




Space and time average model




Average liquid velocity profile for different air flowrate




Main Use :

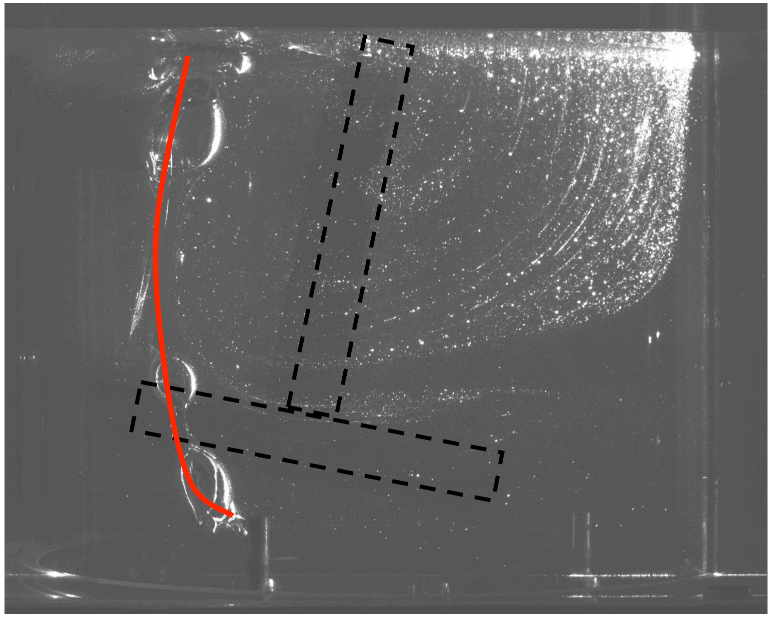
- Have the impact of the gas bubbling on the glass flow and on the free surface

## CFD of the vitrification processes : gas bubbling + mechanical

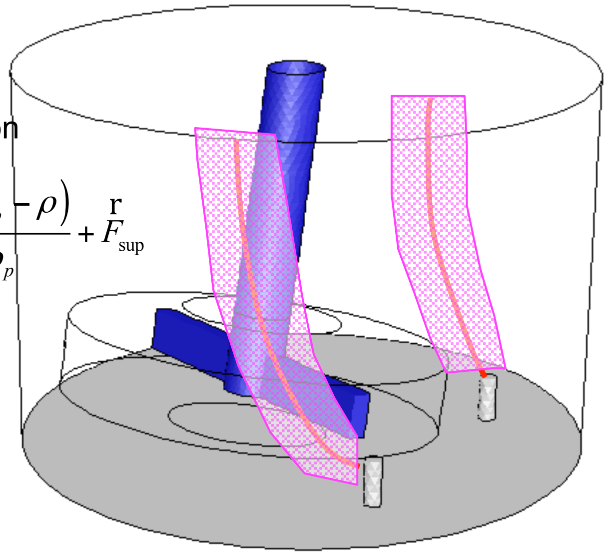
- Mechanical stirring deviates the bubble path
  - Computation of the bubble path with a Lagrangian equation
  - Application of the model along the trajectory

**R. RIVA *(CEA / Grenoble)***

Deviation of the bubble train trajectory



Computation of the bubble train path



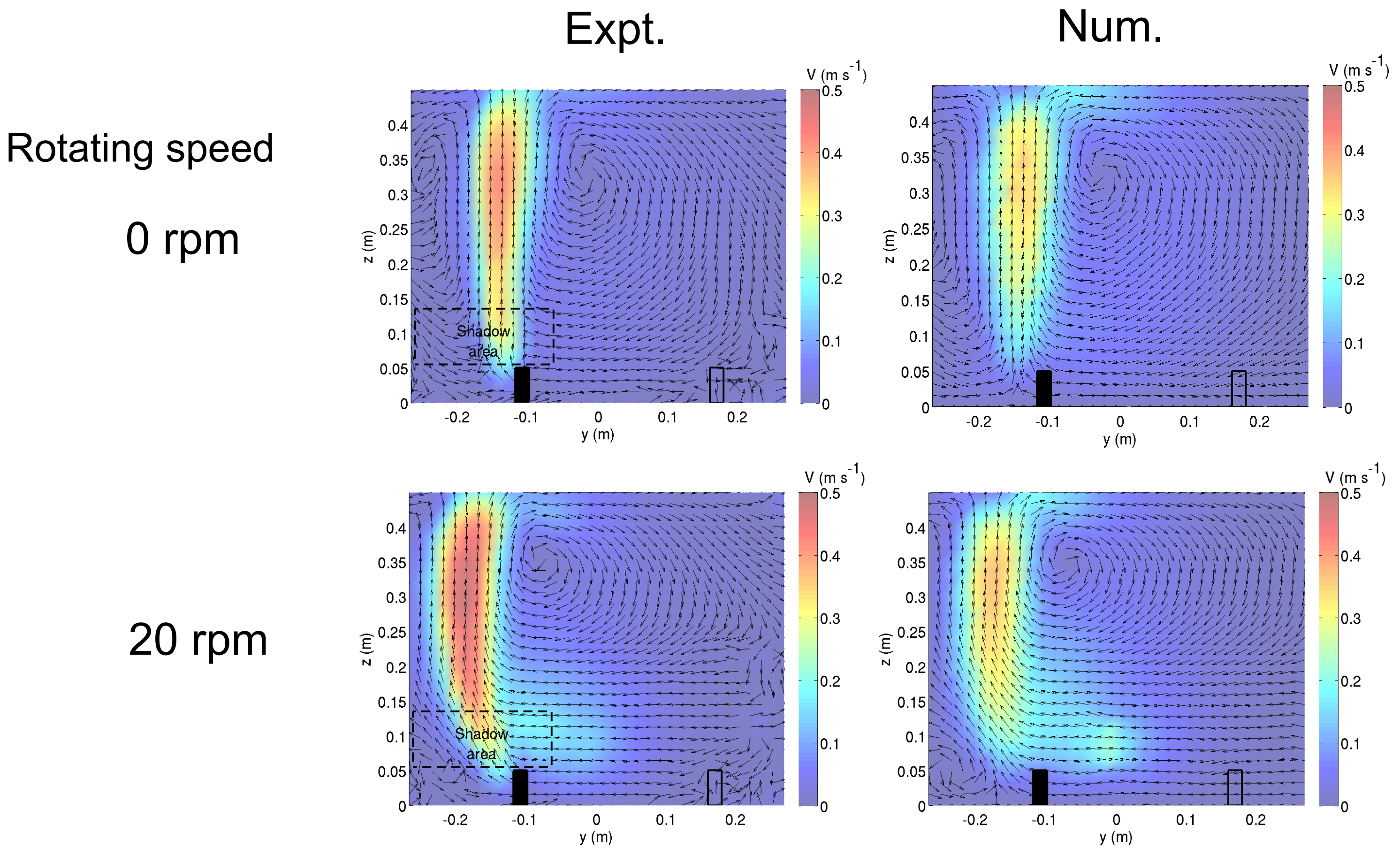
Lagrangian equation for the trajectory

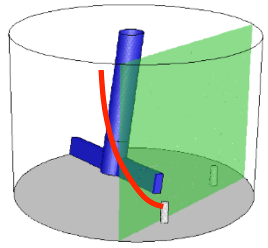
$$\frac{d\vec{u}_p}{dt} = F_D\left(\vec{u} - \vec{u}_p\right) + \frac{\vec{g}\left(\rho_p - \rho\right)}{\rho_p} + \vec{F}_{sup}$$

Main Use :

- Description of bubble trajectory

## CFD of the vitrification processes : gas bubbling + mechanical



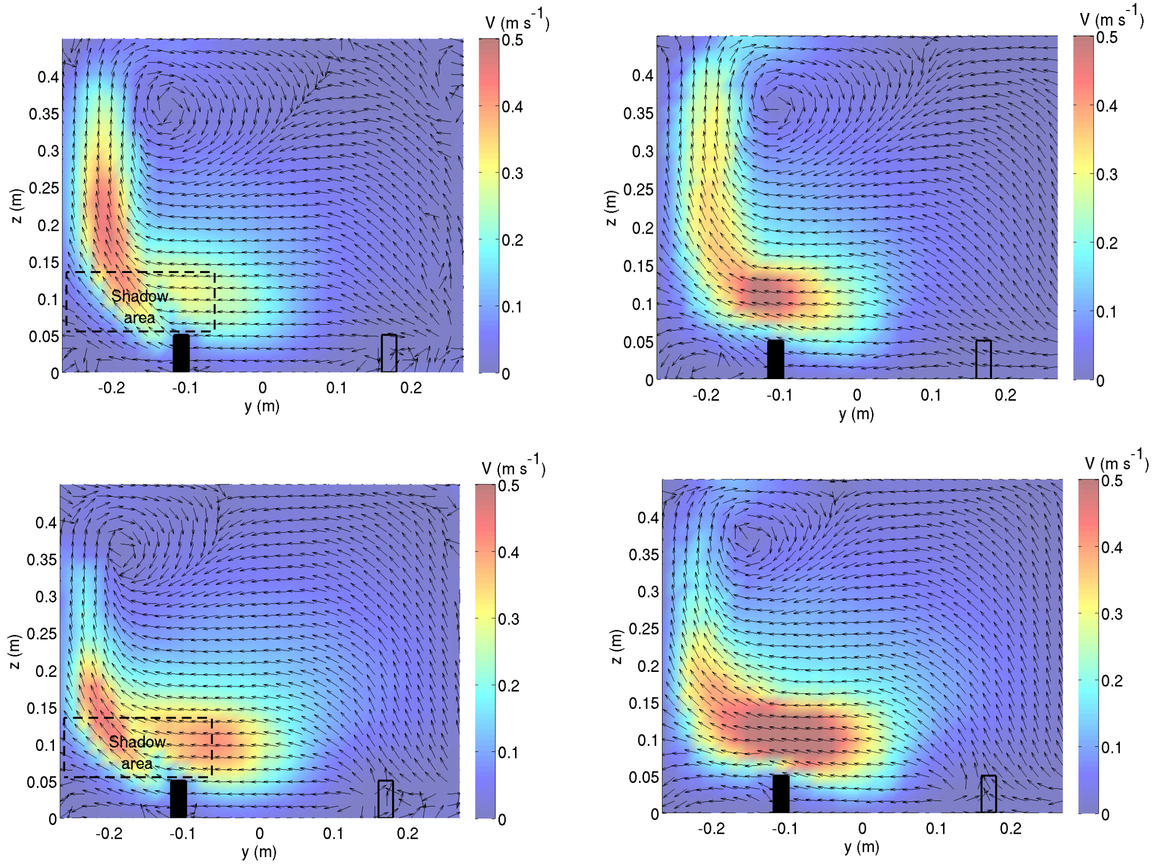
## CFD of the vitrification processes : gas bubbling + mechanical

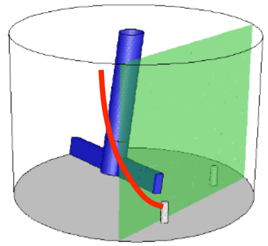
Expt. | Num.

Stirring speed

40 rpm

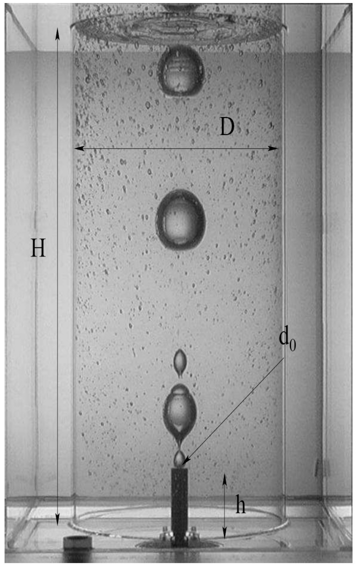
60 rpm



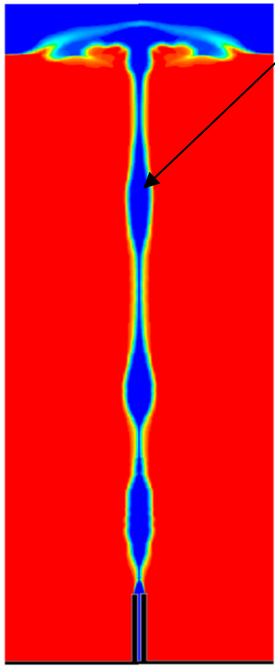



## CFD of the vitrification processes : gas bubbling

- Direct simulation of gas bubbling
  - Volume Of Fluid (VOF) model is use
  - Numerical excitation is needed to obtain separated bubble flow



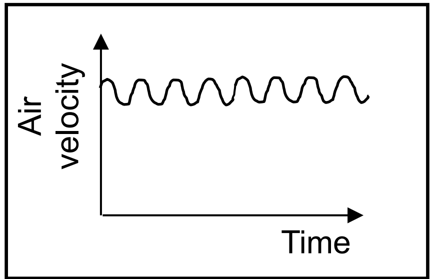

Problem : No bubbles are formed

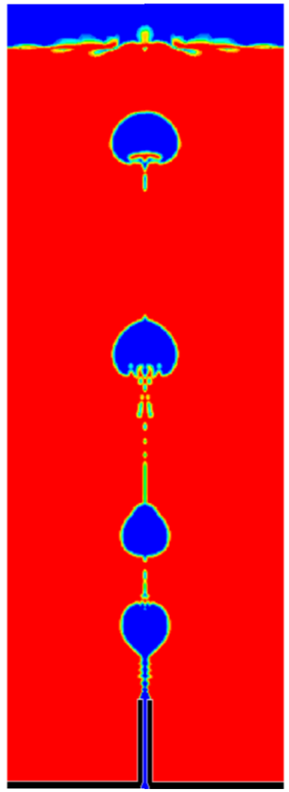


Air inlet

Air jet is a Rayleigh-Plateau instability. This instability should be numerically excited.

With high frequency modulation of the inlet air flow (300 Hz and 4% magnitude)

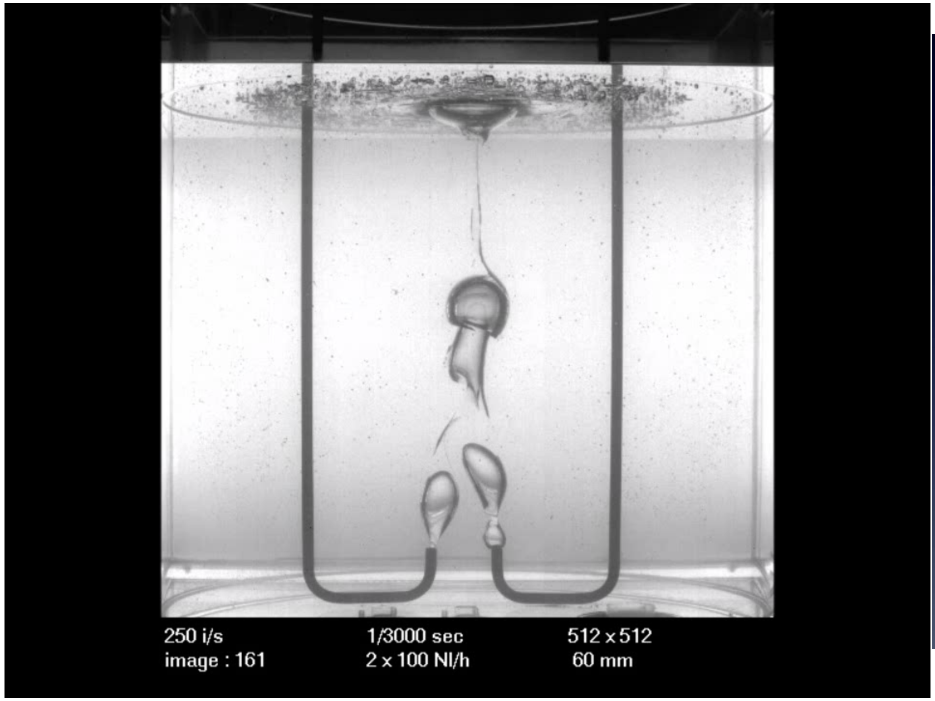


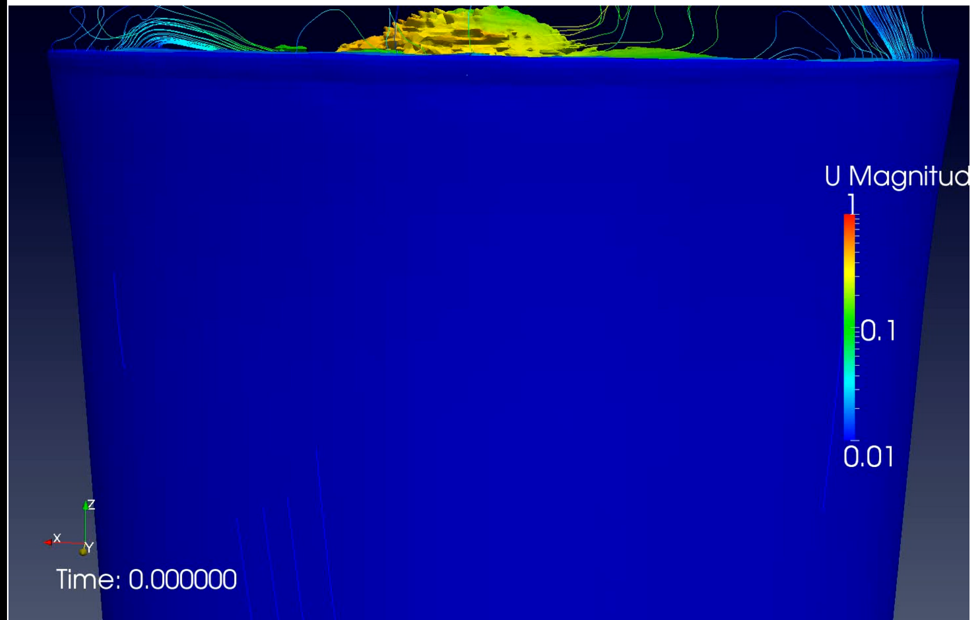



# 2 – Computational Fluid Dynamics (7/7)

Complex diphasics flows have been successfully simulated.

Slow motion (real time / 10)




Main Use :

- Study the impact of the bubble formation on the water-cooled bubblers
- Find optimal position of the bubblers to enhance the convection

# 2 – Computational Fluid Dynamics (7/7)

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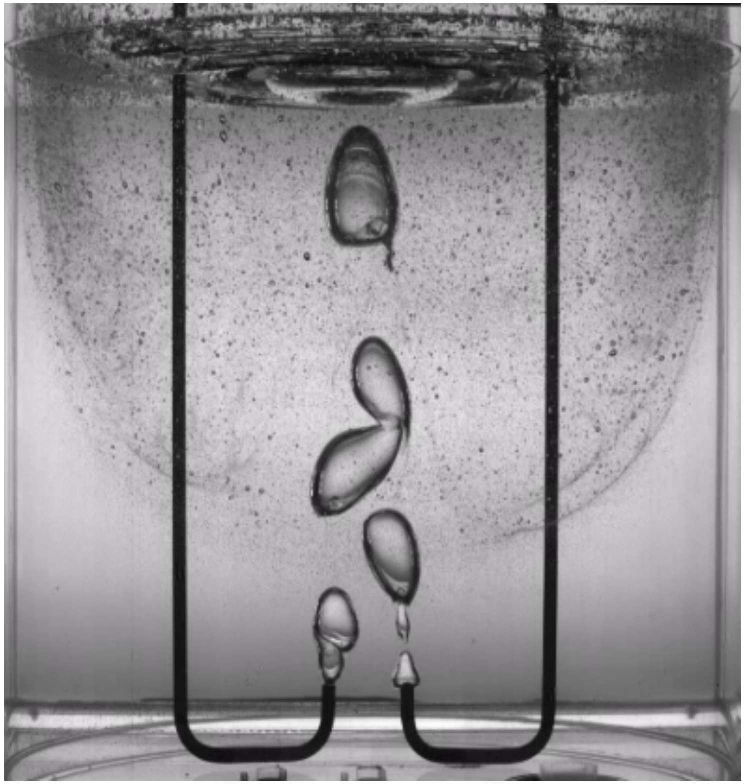
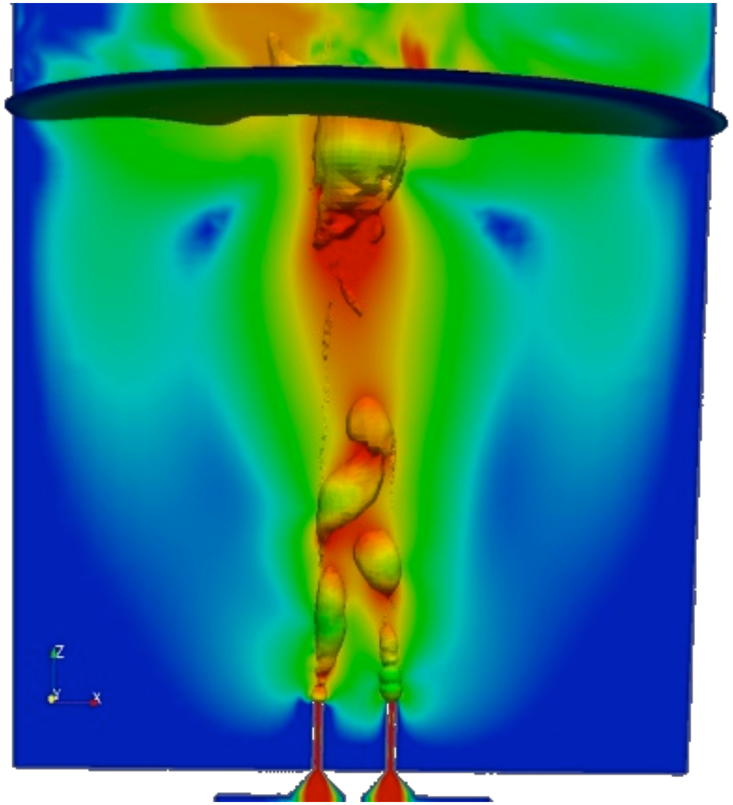
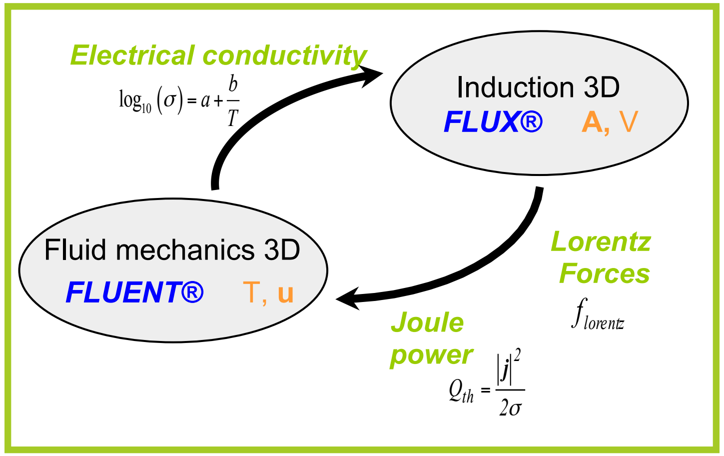


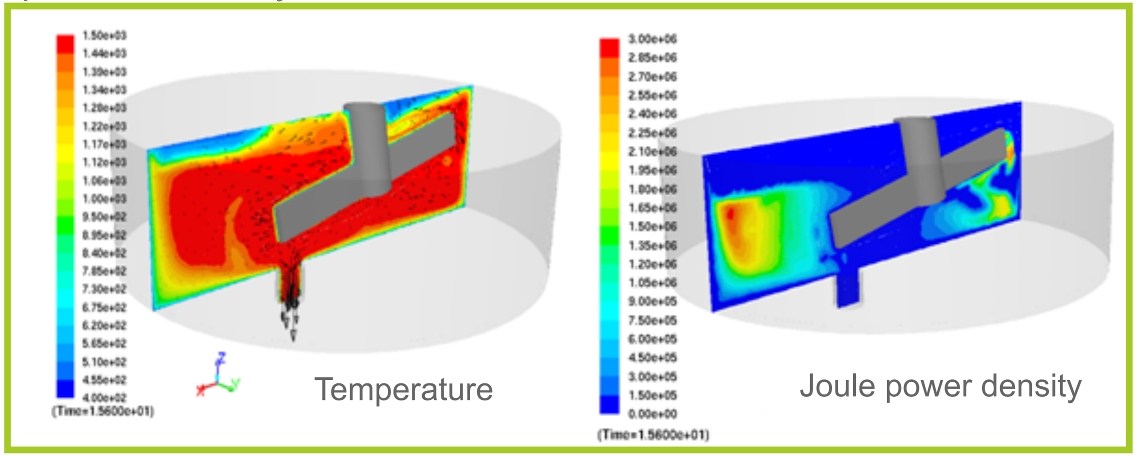


Main Use :
- Study the impact of the bubble formation on the water-cooled bubblers
- Find optimal position of the bubblers to enhance the convection

## Coupling the physics

- First coupled simulation between induction, CFD and thermal simulation of molten glass with Flux-Expert in 1992. All computations were axisymetric.
- Since 2004, files exchange between Fluent and Flux (3D) software
  - Temperature, Joule power density, Lorentz forces
- Recent work : induction equations solved in the CFD software (Openfoam or Fluent) with a finite volume formalism.
  - Use of specific boundary conditions

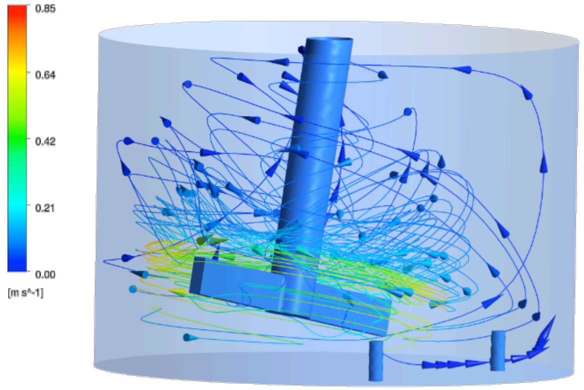


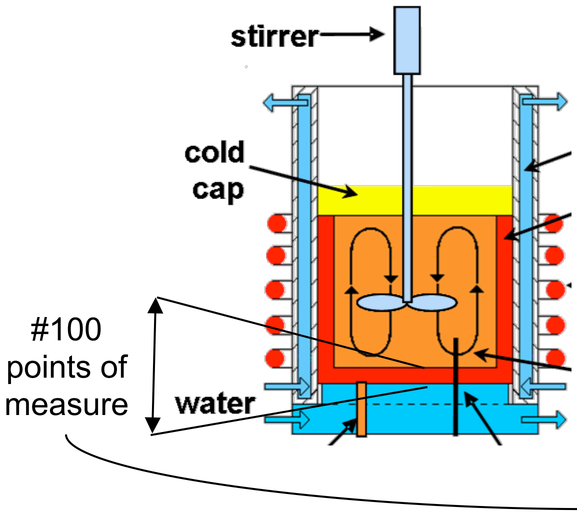



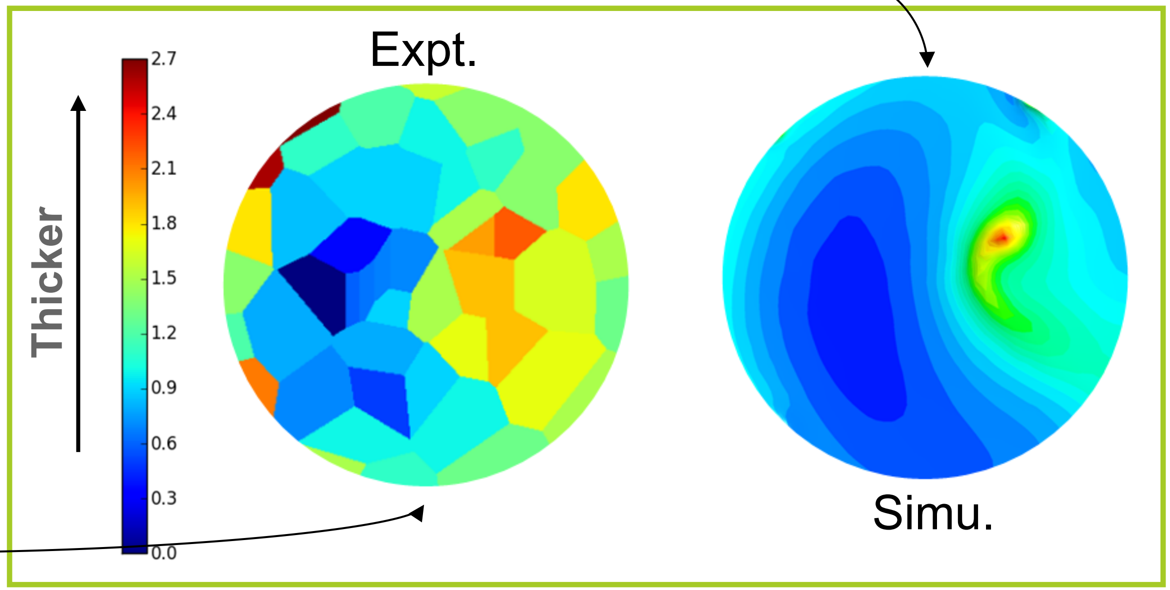
## Validation example :

## Skull melter thickness at the bottom of the crucible

- Inactive glass
- One week long experiment
- Good agreement between simulation and expt. data
- Validation of the fluid flow simulation



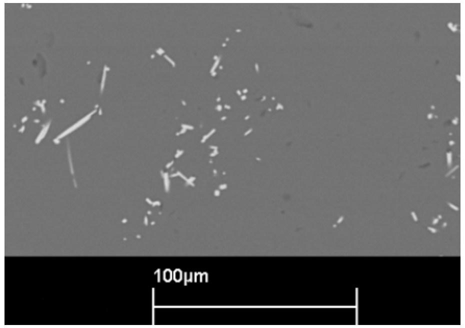



# 2 - NOBLE METAL PARTICLES (1/5)

## Noble metal particles

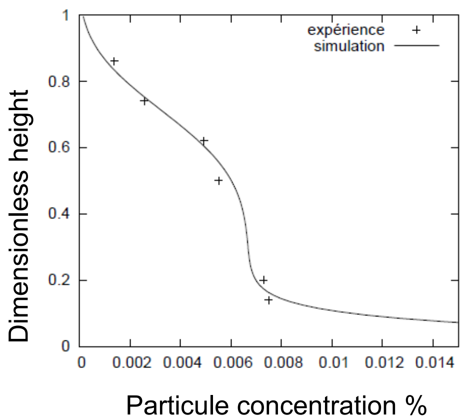
- In the nuclear glass, noble metal particle are present
  - Concentration depending on the waste type (0.1 to 3 %w)
  - Ruthenium dioxide ($RuO_2$) needles
  - Palladium ($Pd$) spheres
- Non negligible impact on CFD and induction

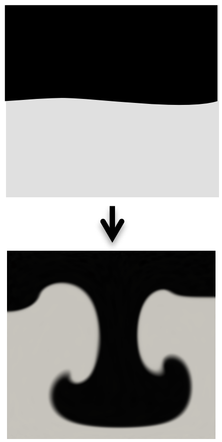
SEM picture of simulated nuclear glass



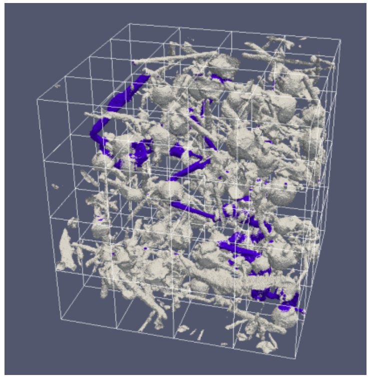

### CFD : Sedimentation phenomena

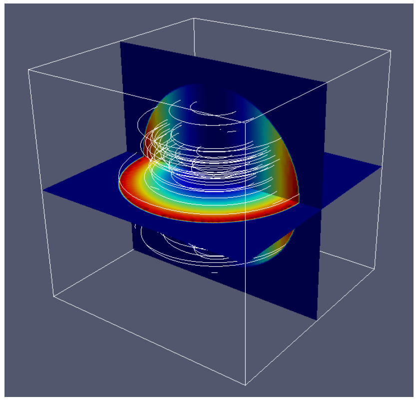
- Individual settling
- Cloud settling



### Induction : Effective electrical conductivity





## Noble metal particles settling

- PHD work G. Barba Rossa

### Individual settling (Eulerian model)

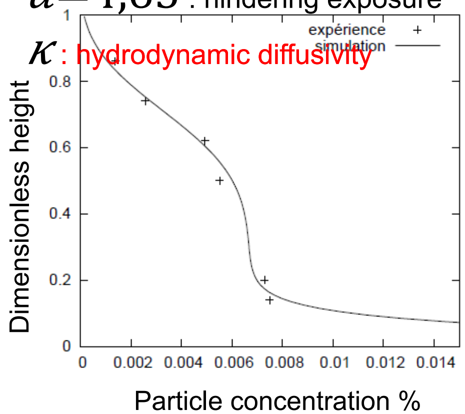
$$\partial C/\partial t + \mathbf{u}\cdot\nabla C + \nabla\cdot\left(\tau \mathbf{g}\, C\left(1 - C/C_a\right)^{\alpha}\right) = \nabla\cdot(\kappa \nabla C)$$

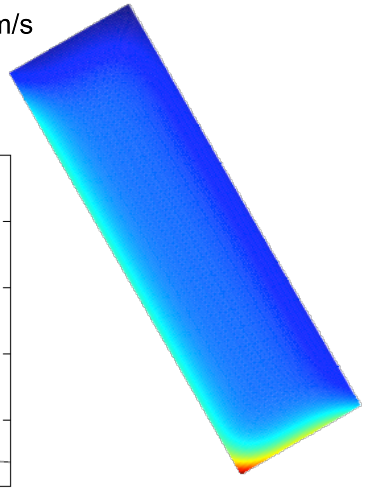
$\tau$ : Stokes terminal velocity ~ 1µm/s

$C_a$ : maximum concentration

$\alpha = 4{,}65$ : hindering exposure

$\kappa$ : hydrodynamic diffusivity

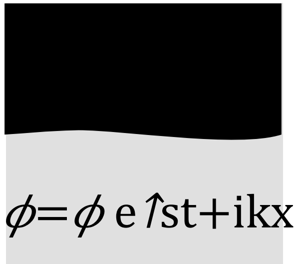


1- Puig J, Penelon B, Marchal P, Neyret M (2014) Rheological Properties of Nuclear Glass Melt Containing Platinum Group Metals. Proc. Mat. Sci. 7:156-162.

### Cloud settling through Rayleigh-Taylor instabilities

Density of glass depend on the particles concentration :

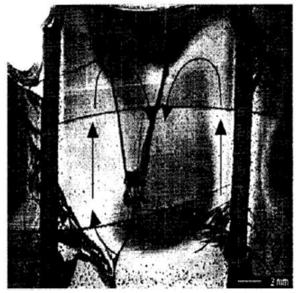


$$\phi = \phi\, e^{st+ikx}$$

$$\rho(C) = \rho_f\,(1 + \beta_p C)$$

$$\beta_p = \rho_p/\rho_f - 1 \approx 2{,}1$$

$s > 0$

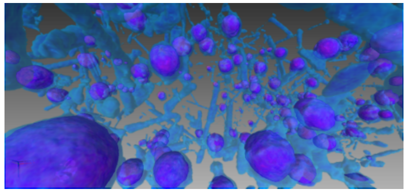


Klouzek et al.

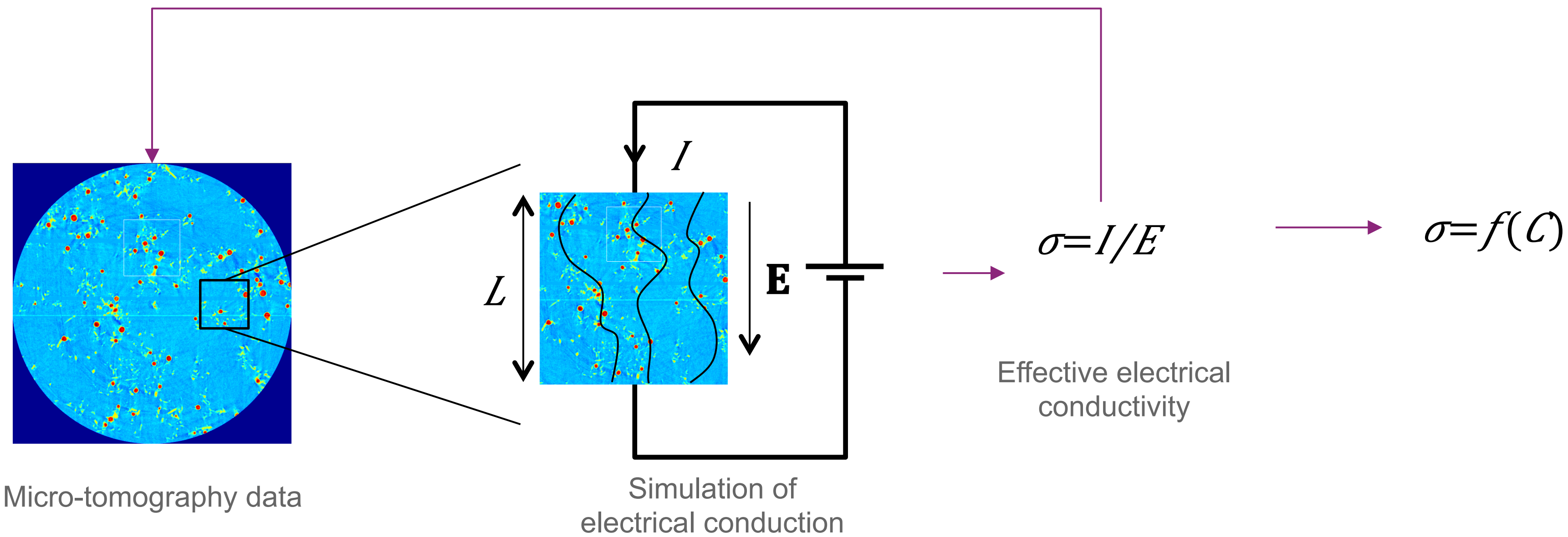
$$s_{max}^{-1} \propto \eta^{1/3} \left(\beta_p \Delta C\right)^{-2/3} \sim 1\ \mathrm{s}$$

## Noble metal particles effect on electrical conductivity

- Micro-tomography X of glass sample
- Simulation of electrical conduction in small parts of the sample
    - Low conductivity for the glass
    - High conductivity for the particles
- Law of effective electrical conductivity function of the concentration



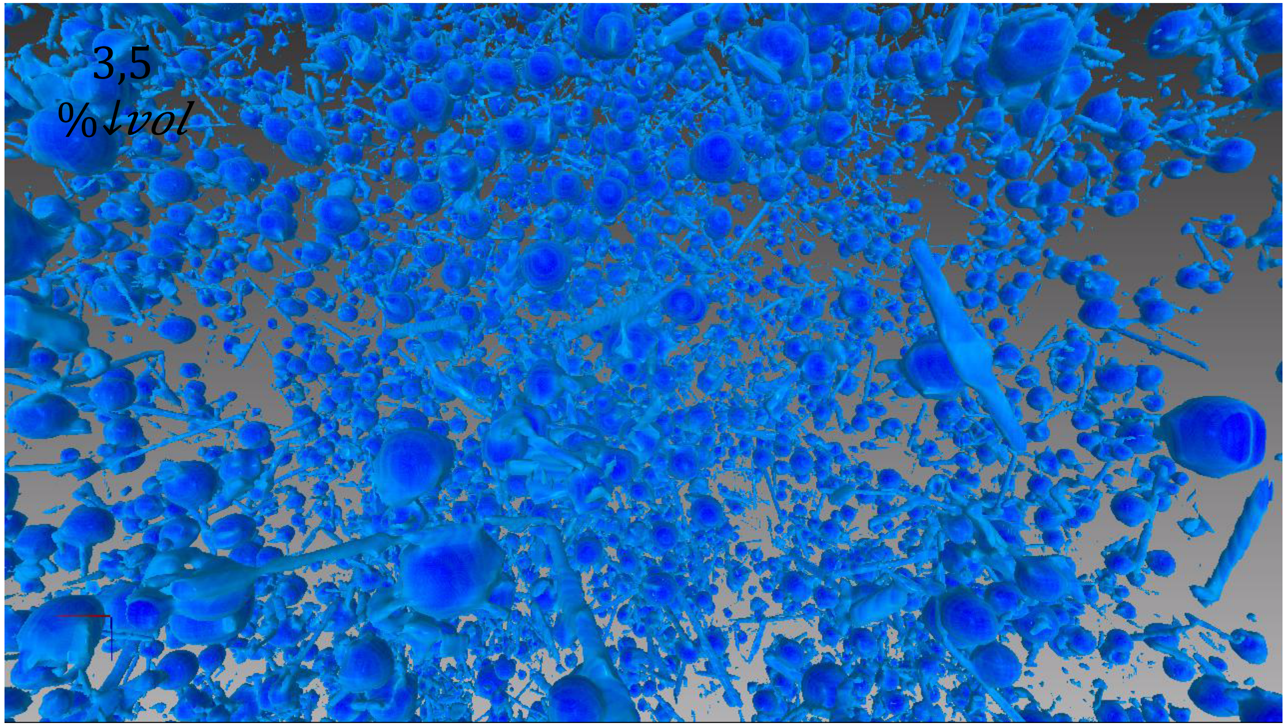
Effective electrical conductivity « measurement »



# 2 - NOBLE METAL PARTICLES (4/5)

## X Micro-tomography

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

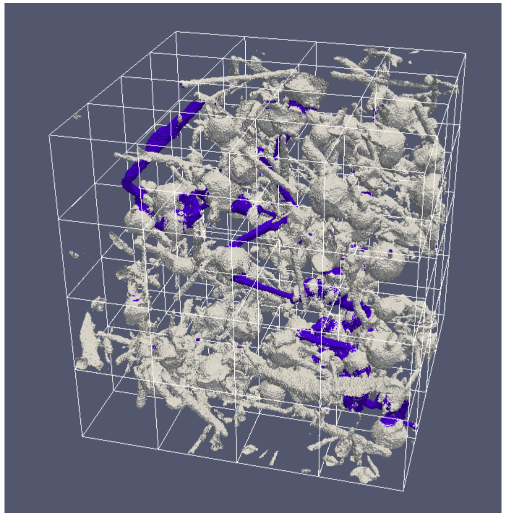


# 2 - NOBLE METAL PARTICLES (5/5)

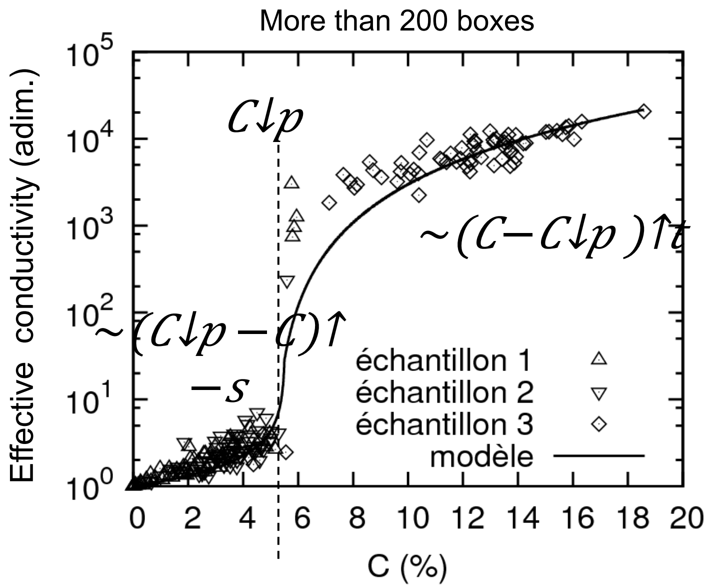
## Noble metal particle effect on electrical conductivity

- Micro-tomography X
- Simulation at low scale

**Example of a computation box**

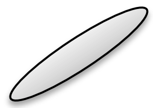


**Percolation curve**




Garboczi et al. 1995

$C_p \approx 0.6/\Psi$



aspect ratio $\Psi \approx 10$

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

# 2 - NOBLE METAL PARTICLES (5/5)

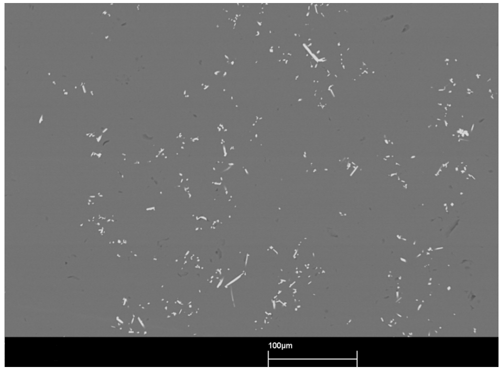
## Multiscale approach -- Application

- Settling modelling coupled with percolation model and induction heating
  - Multi-scale approach

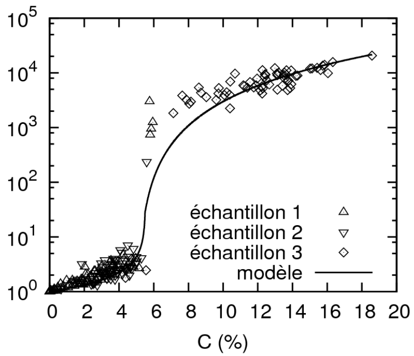
| | $10^{-4}$ m | $10^{-2}$ m | $10^{0}$ m |
|---|---|---|---|

**Expt.**

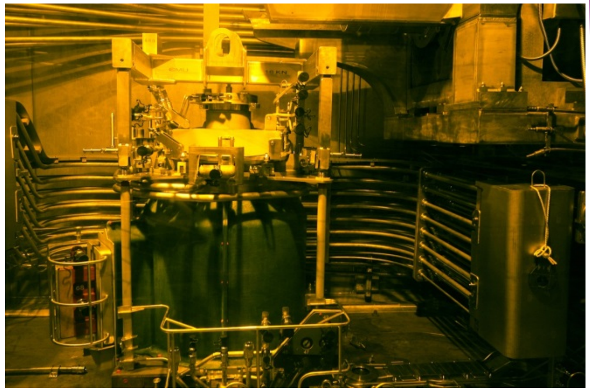
$10^{-4}$ m:




$10^{-2}$ m:

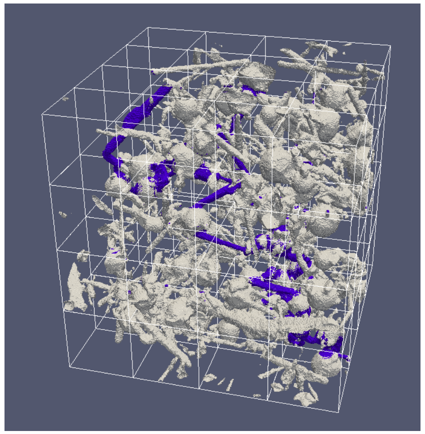



$10^{0}$ m:

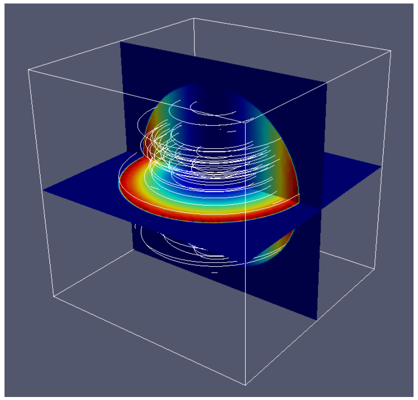


**Simulation**

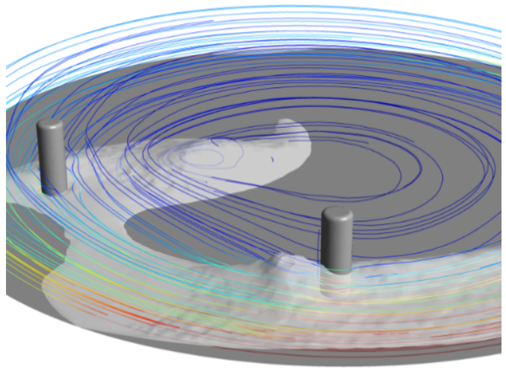
$10^{-4}$ m:



$10^{-2}$ m:



$10^{0}$ m:



# Summary

1 − French radioactive waste vitrification technology presentation

2 - Vitrification simulation

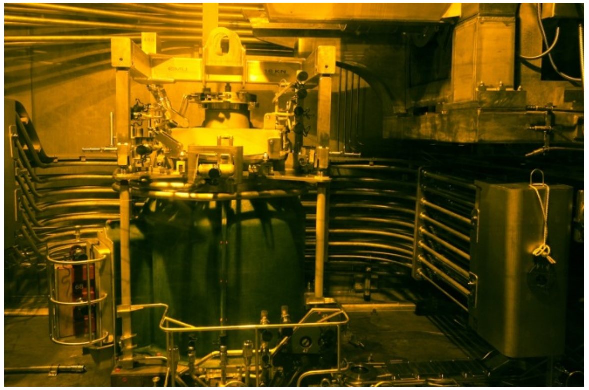
2-a Induction modeling

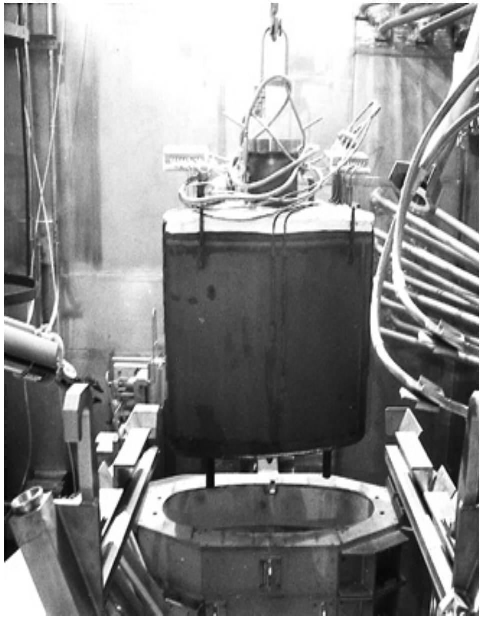
2-b CFD modelling

2-c Coupled physics

3 – Applications and Ongoing development

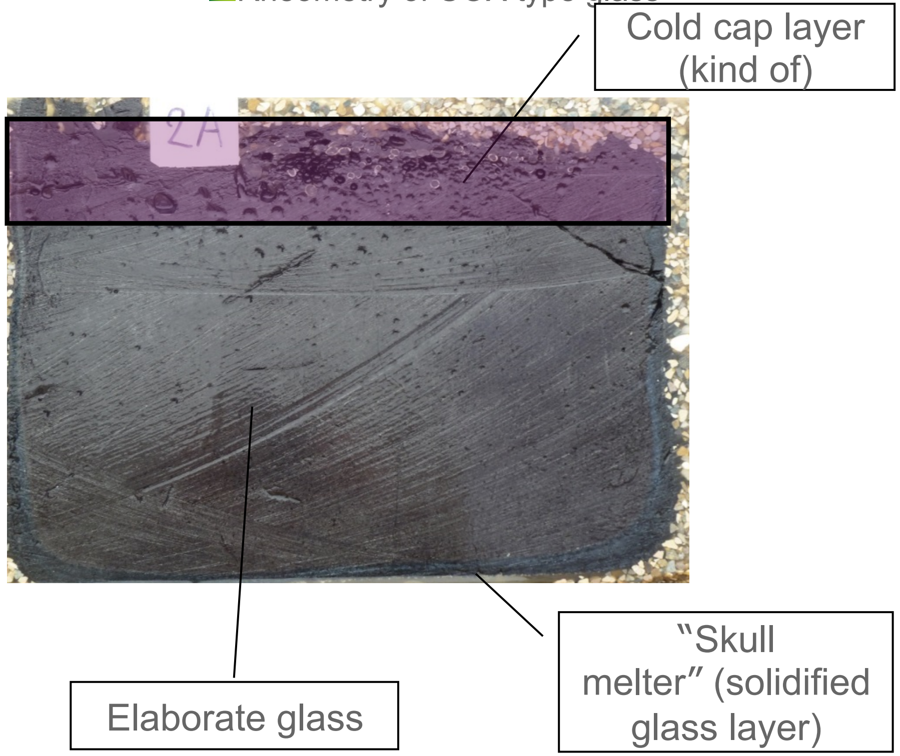
4 - Conclusion



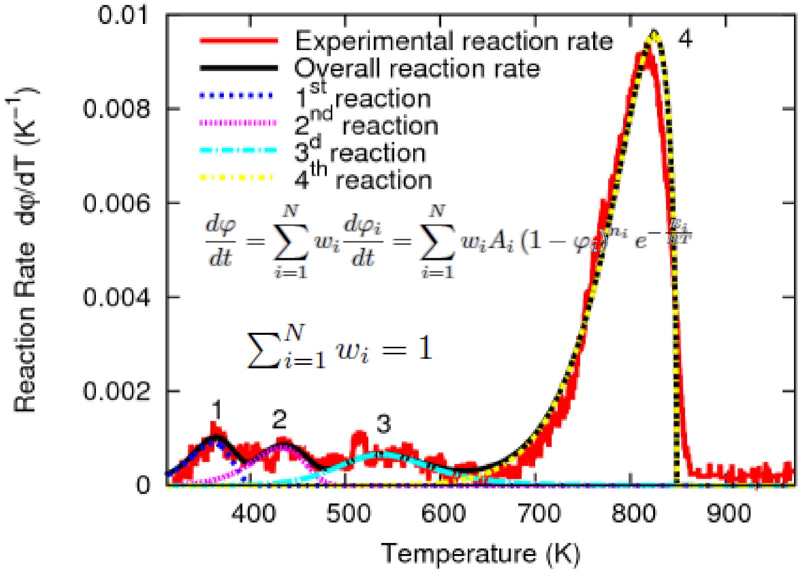


## Simulation of chemical reactions during vitrification process

- Idea : adapt the work of Hrma – Pavel – Pokorny – Dixon (PNNL DOE) on the cold-cap reaction modelisation
- PHD work planned to start in 2018
- Thanks to new characterizations of glass
  - TGA measurement
  - Rheometry of UOX type glass[1]




Chemical reactions in the cold cap layer*




* J. Chun, D. Pierce, R. Pokorny and P. Hrma : Cold-cap reactions in vitrification of nuclear waste glass, Thermochimica Acta, 559 (2013) 32-39

# 4 - Simulation of CCIM vitrification furnace

## Examples of applications

- Use to enhance the comprehension of what happens in the glass
  - Diagnostic tool
  - Determination of residence time of material in the furnace
  - Reducing unwanted electric current
- Design and sizing stage
  - Induction
  - Design of temperature sensors
  - CFD for shape and position of stirrer. Bubblers number and position.
- Industrial operation support

## Potential benefit

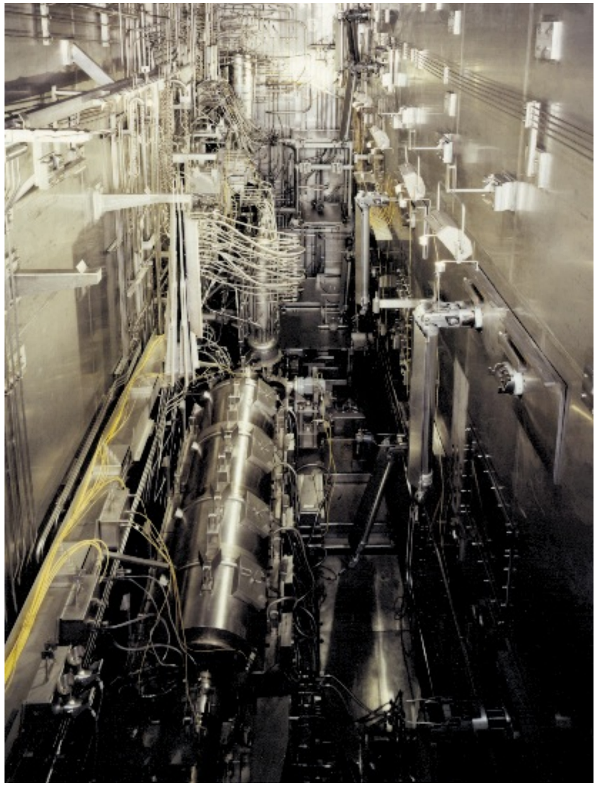
- Enhanced reliability
- Increase of the life time
- Increase of the glass throughput
- Innovative concept

# Conclusion

- The development of numerical simulation has been done since the beginning of the vitrification processes development.
  - They play a role increasingly important in projects.
  - Simulations are validated

- Since 2000, simulations are no more used only for the global conception design
  - Optimization of details design,
  - Comprehension and diagnostic tool,
  - Testing new innovative concepts.

- What restricts the use of numerical simulation ?
  - Computation time
  - Input data knowledge -> Development of the measurement techniques of the physical properties of glass
  - Experimental data for validation or build-up new model -> mock-up or full scale inactive pilot experiment



DE LA RECHERCHE À L'INDUSTRIE

cea

Thank you for your attention

Any questions ?

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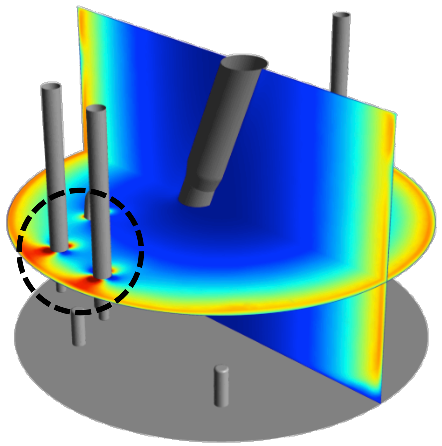
Direction de l'Energie Nucléaire
DTCD
SCDV

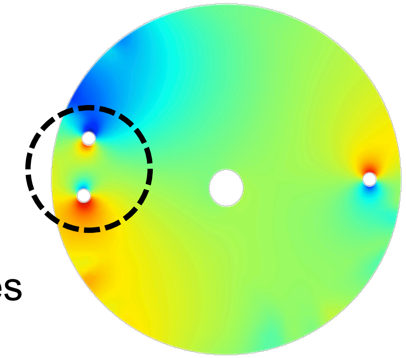
# Réponse Magnéto-Thermo-Hydraulique

- Champ appliqué $B = 1{,}3\ \mathrm{mT}$, $P_J = 181\ \mathrm{kW}$
- Température régulée $T_h = 1473\ \mathrm{K}$



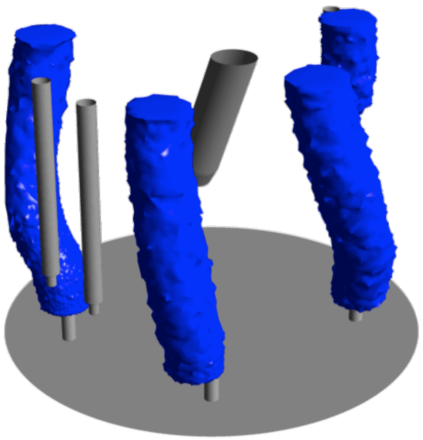
surpuissances locales

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

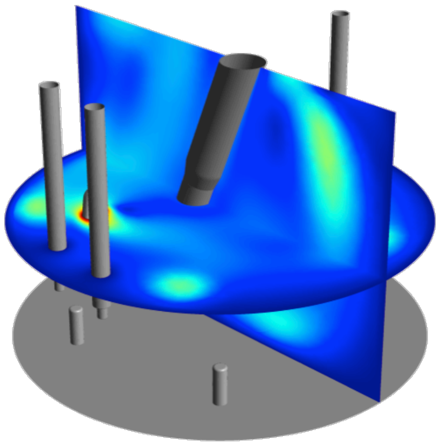


différences de potentiel induites

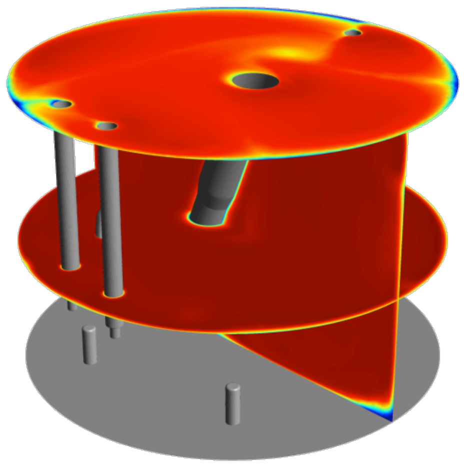
$V_i \in [-5;5]\ \mathrm{V}$



$iso-S_z = 500\ \mathrm{N/m^3}$



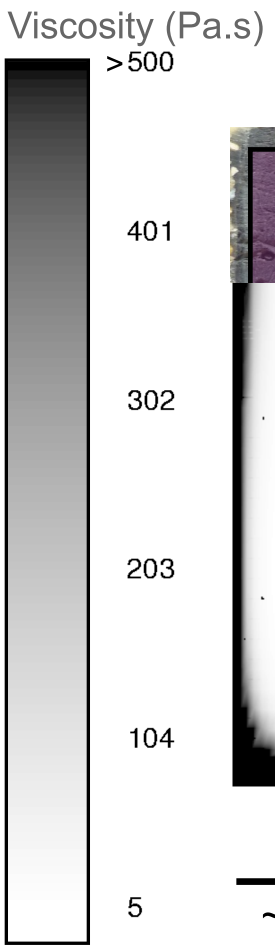
$U \in [0;1]\ \mathrm{m/s}$

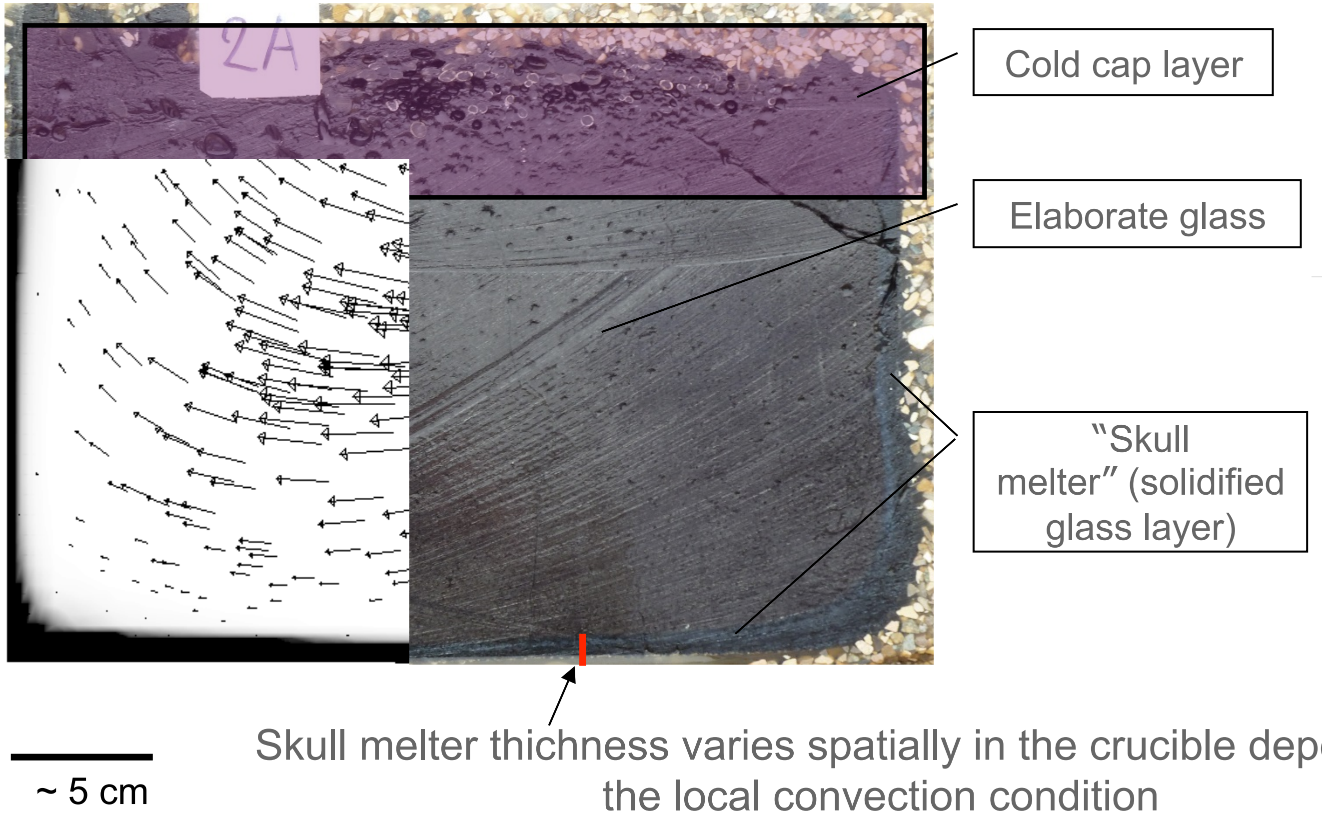


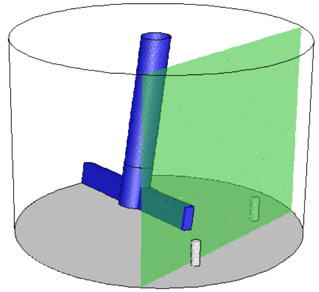
$T \in [1200;1480]\ \mathrm{K}$

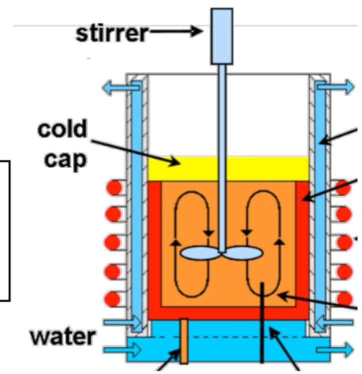
# 3-b Validation exemple (1/2)

Slice of inactive glass in a cold crucible during elaboration. The cold crucible has been removed after fast cooldown






Skull melter thichness varies spatially in the crucible depending on the local convection condition

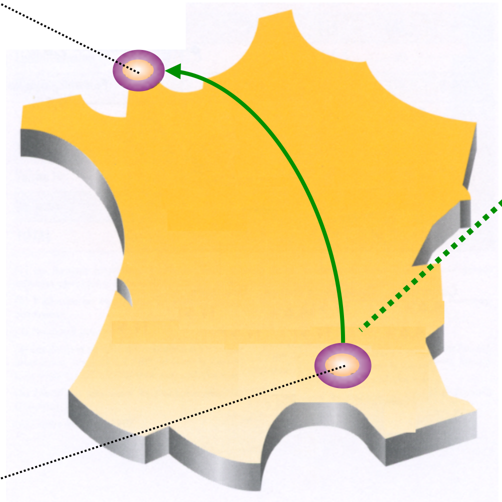
# CEA / AREVA Joint Vitrification Laboratory (LCV)

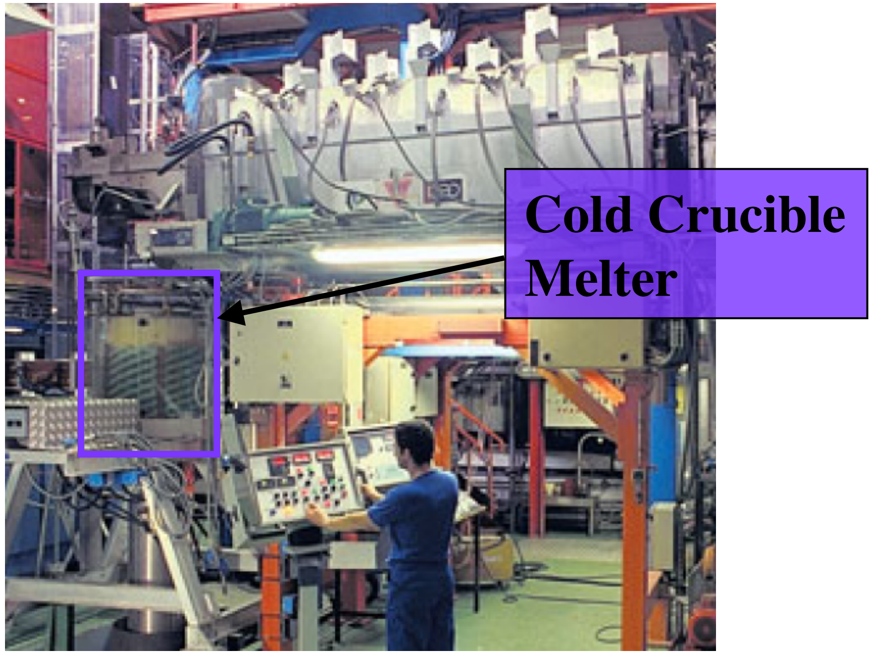
## Areva

- Commercial and industrial operator of 2 vitrification plants


**La Hague**
**(AREVA)**
**Industrial plant**

**Marcoule**
**(CEA)**
**Research center**



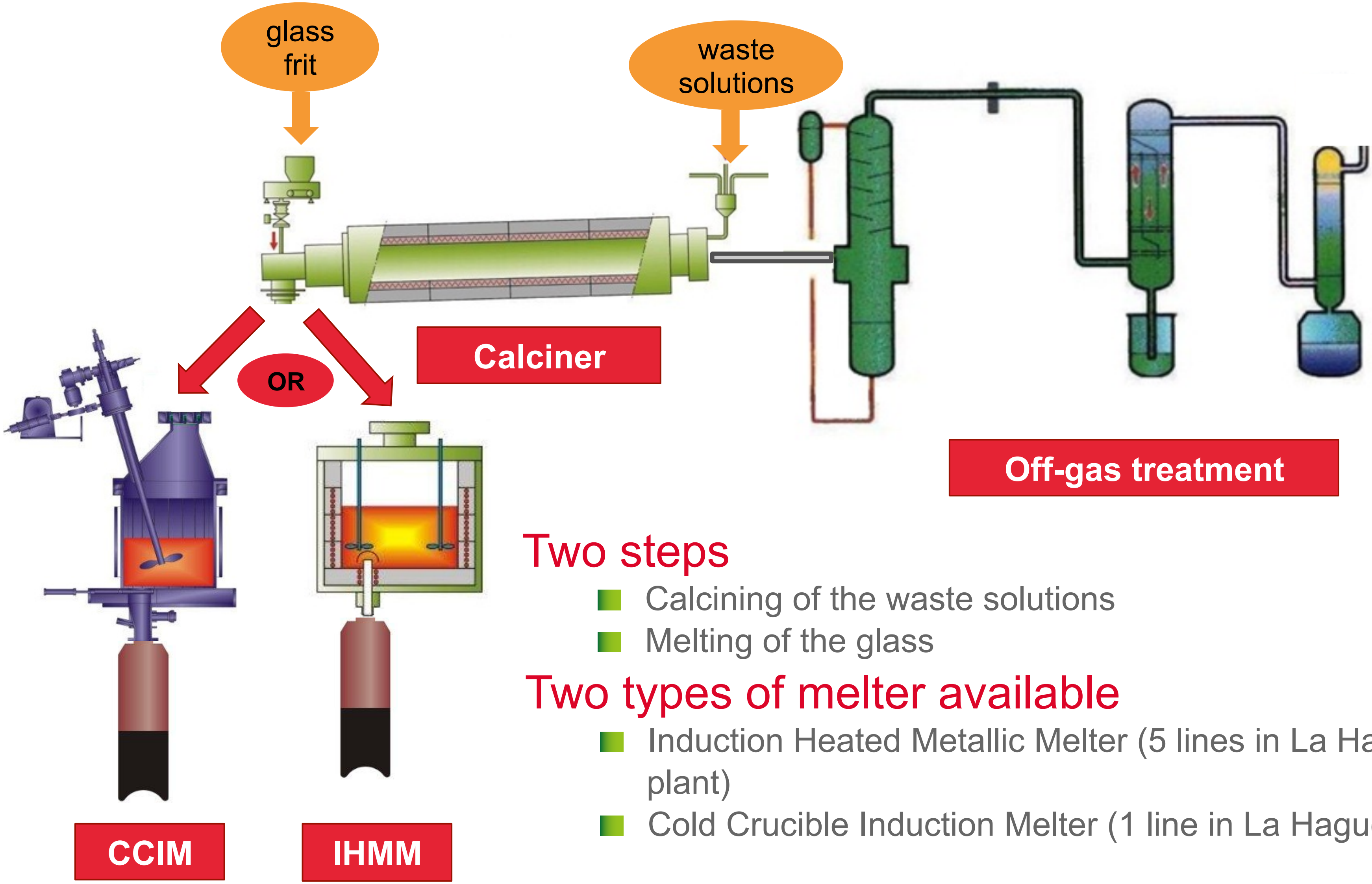



Current pilot of vitrification process
CEA Marcoule (inactive cell)

## French Atomic Energy Commission (CEA)

- Confinement Research Engineering Department
  Waste Confinement and Vitrification Service

# The French vitrification process




## Two steps

- Calcining of the waste solutions
- Melting of the glass

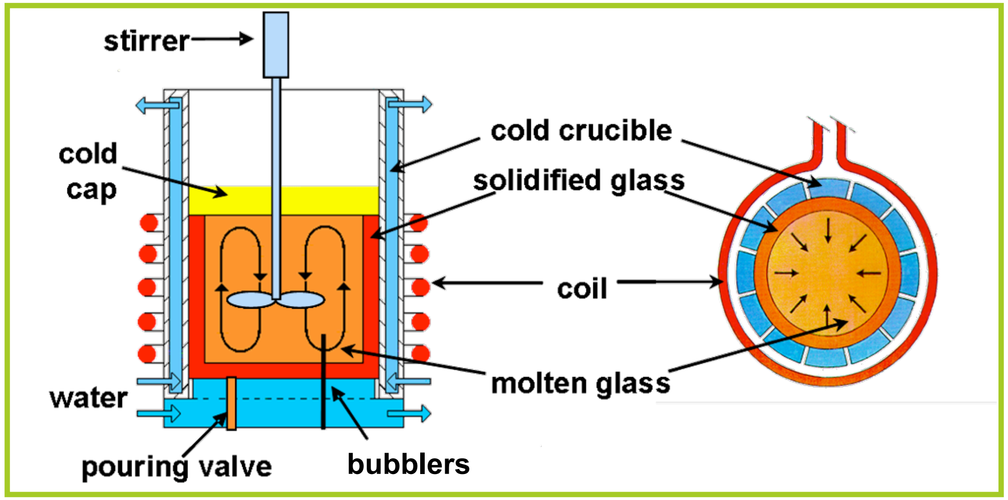
## Two types of melter available

- Induction Heated Metallic Melter (5 lines in La Hague plant)
- Cold Crucible Induction Melter (1 line in La Hague plant)

# Purpose of the numerical simulation

## Challenges of the numerical simulation for CCIM

- Inductor connected to a high-frequency current generator
- Cooling of the metal walls
- Formation of a skull-melter
- Mechanical stirrer and air bubbling




## Issues

- Complex geometry
- Many physics are involved
  - Direct induction
  - Forced and natural convection
  - Radiative heat transfer
- Temperature dependence of the physical properties

| Physical properties | Unity | 500 K | 1500 K |
|---|---|---|---|
| Electrical conductivity | $\Omega^{-1}.m^{-1}$ | $10^{-5}$ | 20 |
| Dynamic viscosity (μ) | $Pa.s$ | $10^{14}$ | 1 |
| Specific heat ($C_p$) | $J.kg^{-1}.K^{-1}$ | 900 | 1500 |
| Thermal conductivity (λ) | $W.m^{-1}.K^{-1}$ | 1 | 6 |
| Density (ρ) | $kg.m^{-3}$ | 2850 | 2750 |

## ➲ CCIM Deployment at La Hague / main stages

- **2010** First active operation of the CCIM implemented in the R7 facility
- **2009** Commissioning and inactive tests of the CCIM implemented in the R7 facility
- **2008** Industrialization phase 4. Nuclearized and industrialized CCIM construction for implementation in the R7 facility
- **2006** Industrialization phase 3. Nuclearized CCIM prototype construction for process and glass quality qualification
- **2004** AREVA NC decision to implement a CCIM in R7
- **2000** Industrialization phase 2. Specific CCIM prototype for UMo vitrification
  650 mm in diameter CCIM
  3000 hours of inactive test / industrial capacity
- **1992** AREVA NC decided to study the implementation of the cold crucible
- **1985** Industrialization phase 1. Larger CCIM construction (550 mm in diameter)
- **1981** First CCIM prototype (350 mm in diameter)