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Numerical simulations in the development of the French radioactive waste vitrification processes using induction furnace

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

Trieste (Italy), November 6 - 10, 2017



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

PRESENTATION BY EMILIEN SAUVAGE

EMILIEN.SAUVAGE@CEA.FR



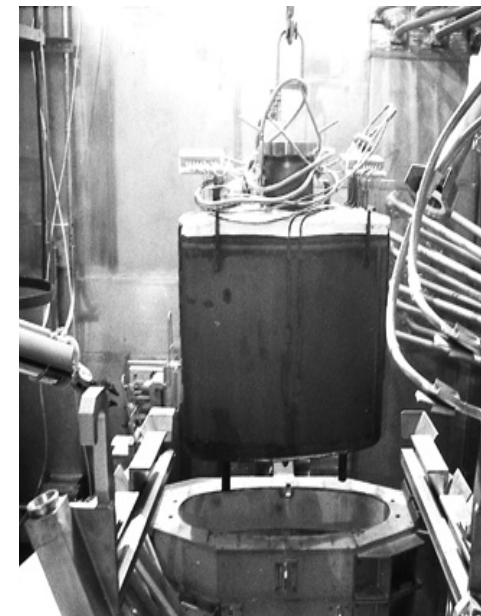
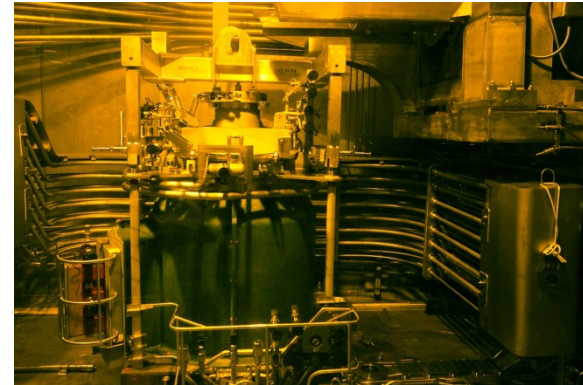
E. SAUVAGE, P. BRUN, A. BONNETIER, G. BARBA
ROSSA, R. DIDIERLAURENT

CEA, DEN, DE2D, SEVT, LDPV

BP 17171, F-30207 BAGNOLS-SUR-CEZE, FRANCE

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



- 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

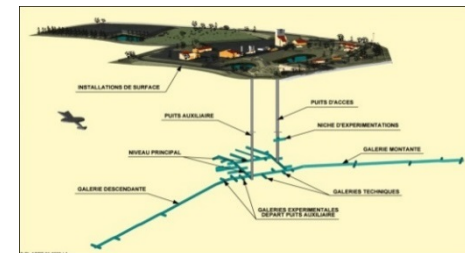
- ✓ Around 20 000 canisters produced (400 kg of nuclear glass each)

- 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

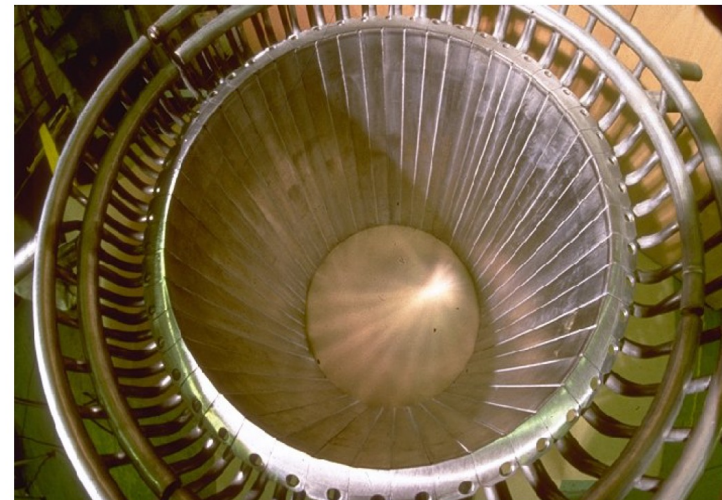


- Need of higher throughputs and higher temperature : in 2004 AREVA NC decision to implement a CCIM in R7

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



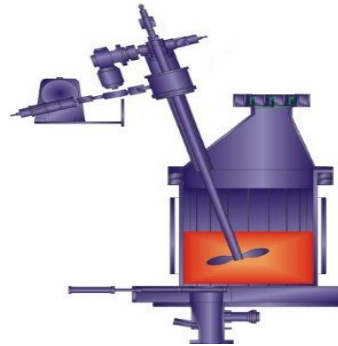
- AREVA has replaced one existing Induction Heated Metal Melter (IHMM) in a production line in the R7 facility at La Hague plant by a Cold Crucible Induction Melter (CCIM)
- April 2010 → Hot operation for the first time ever at the La Hague R7 vitrification facility
- CCIM in commercial operation for 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)



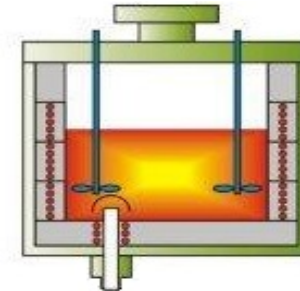
VITRIFICATION PROCESS OPERATED IN THE LA HAGUE PLANT

Furnaces technology overview

Cold Crucible Inductive Melter



Inductive Hot Metallic Melter



Characteristics:

- Induction heating type
- Number in LH plant
- Commissioning in

direct

1

2010

indirect

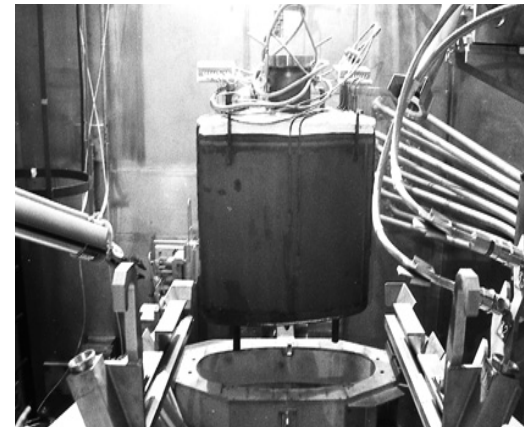
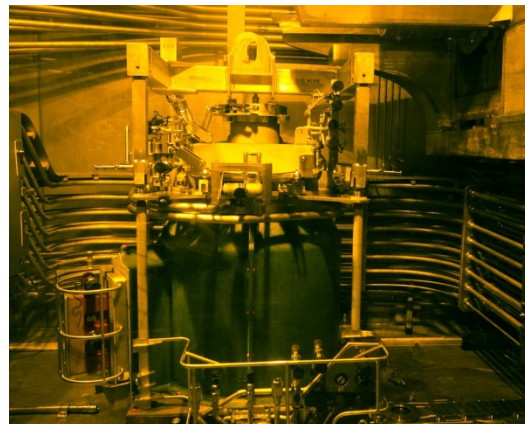
5

1989

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

Oxides	Average composition of industrial glass (w %)
SiO ₂	45,6
B ₂ O ₃	14,1
Al ₂ O ₃	4,7
Na ₂ O	9,9
CaO	4
Fe ₂ O ₃	1,1
NiO	0,1
Cr ₂ O ₃	0,1
P ₂ O ₅	0,2
Li ₂ O	2
ZnO	2,5
Oxides (PF+Zr+actinides) + Suspension of fines Actinide oxides	17
SiO ₂ +B ₂ O ₃ +Al ₂ O ₃	64,4

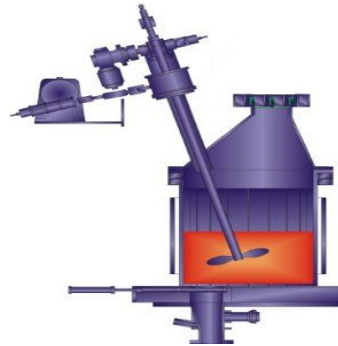
Picture in hot cell
(Areva La Hague)



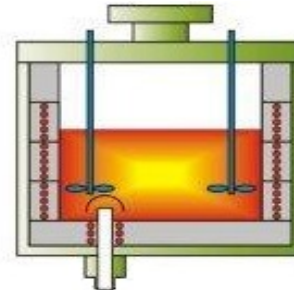
VITRIFICATION PROCESS OPERATED IN THE LA HAGUE PLANT

Furnaces technology overview

Cold Crucible Inductive Melter



Inductive Hot Metallic Melter



IHMM



Characteristics:

- 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

Inside view in CCIM (inactive Marcoule)



Summary

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2-a Induction modeling

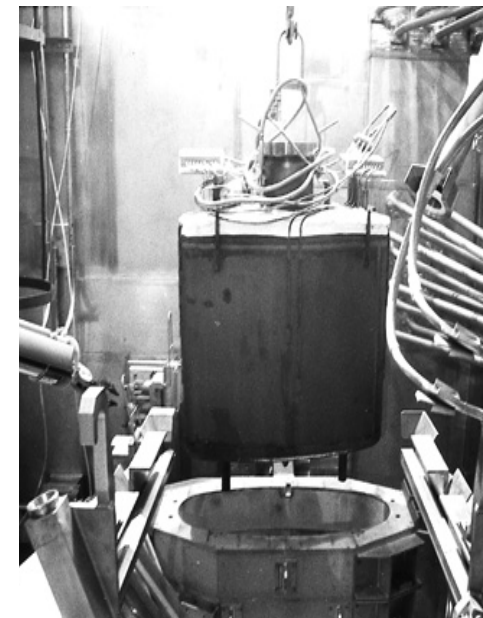
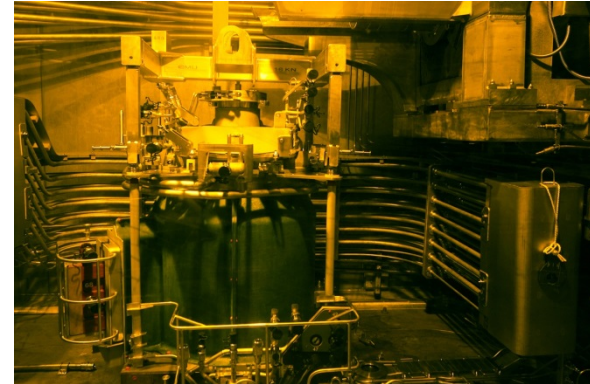
2-b CFD modelling

2-c Coupled physics

2-d Noble metal particles

3 – Ongoing development

4 - Conclusion

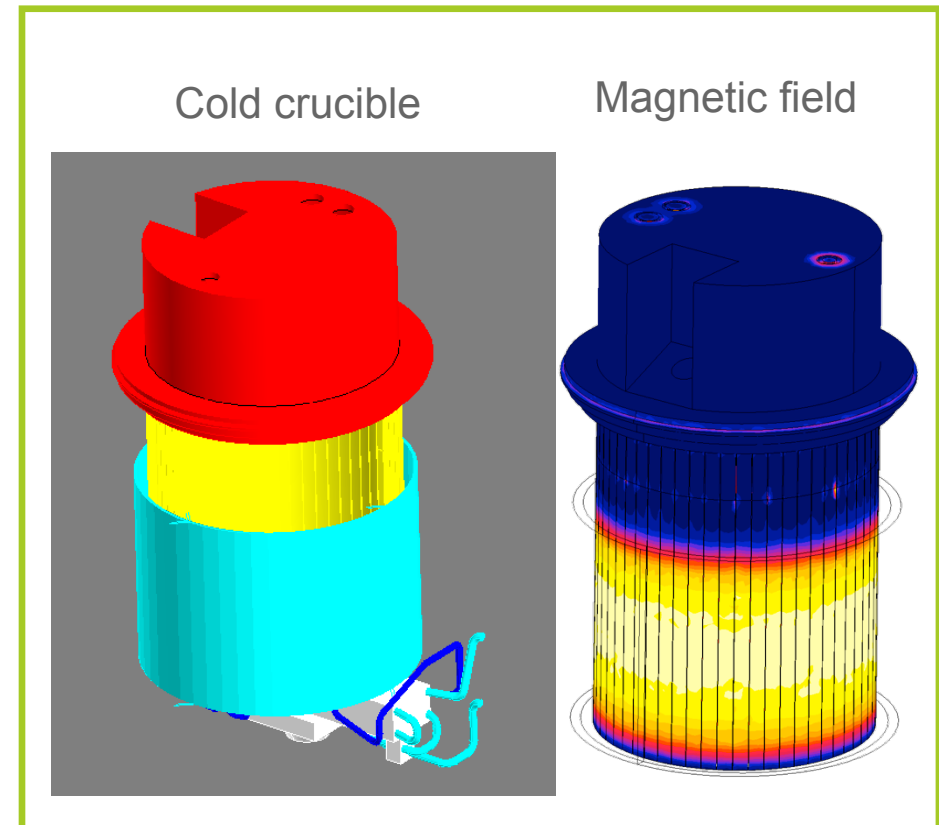


Simulation of induction heating in vitrification processes

- 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



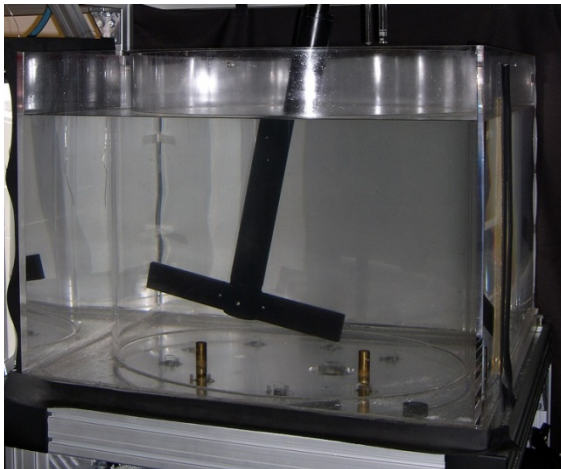
Main Use :

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

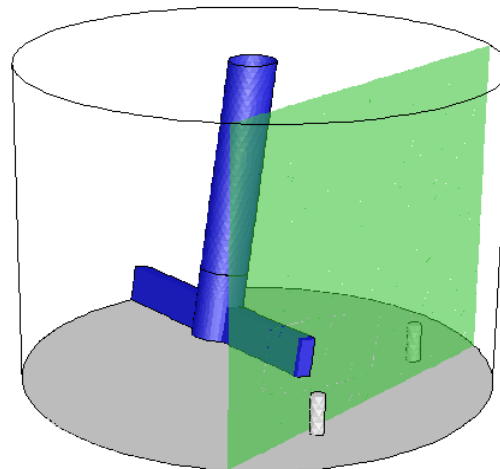
CFD of the vitrification processes : mechanical stirring

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

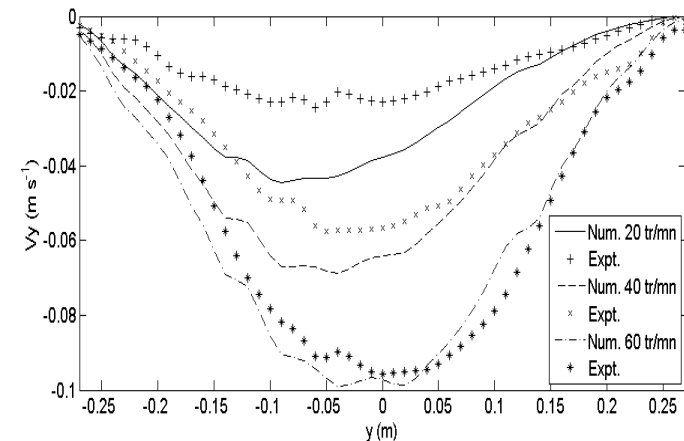
Similarity experiment



3D CFD computing (sliding mesh)



Velocity profile for different rotating speed



Main Use :

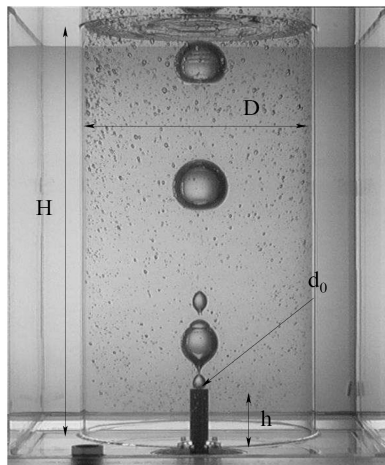
- Shape and position of stirrer, optimised rotating speed

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

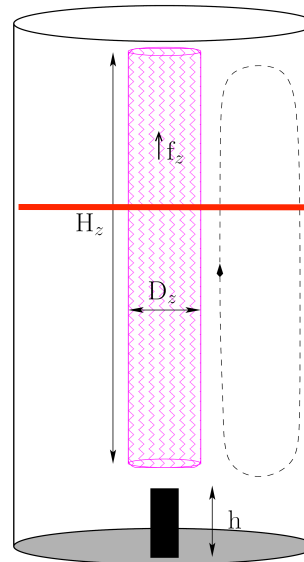
Experiment in hydraulic similarity with silicon oil

R. RIVA (CEA / Grenoble)

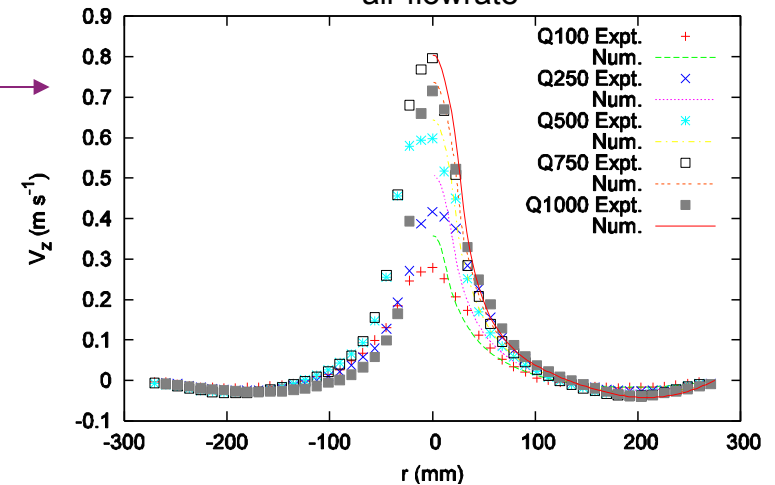


30 to 60 cm

Space and time average model



Average liquid velocity profile for different air flowrate



Main Use :

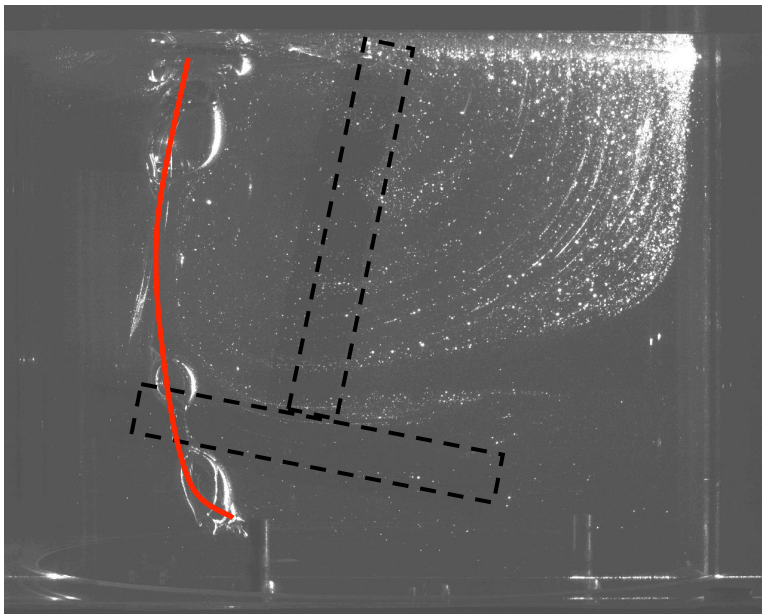
- Have the impact of the gas bubbling on the glass flow and on the free surface

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



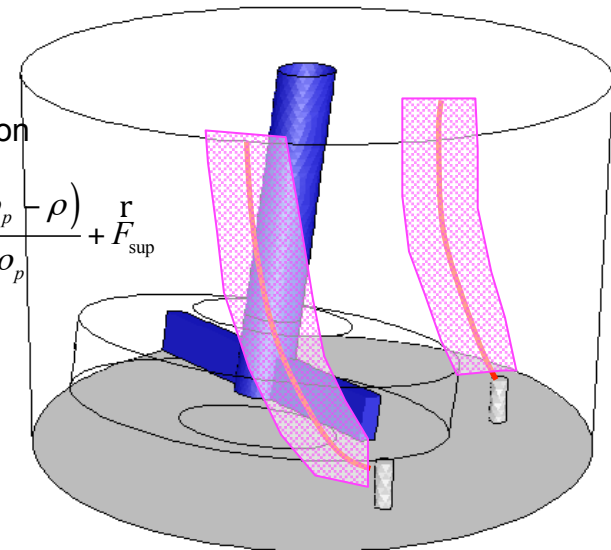
Main Use :

- Description of bubble trajectory

Computation of the bubble train path

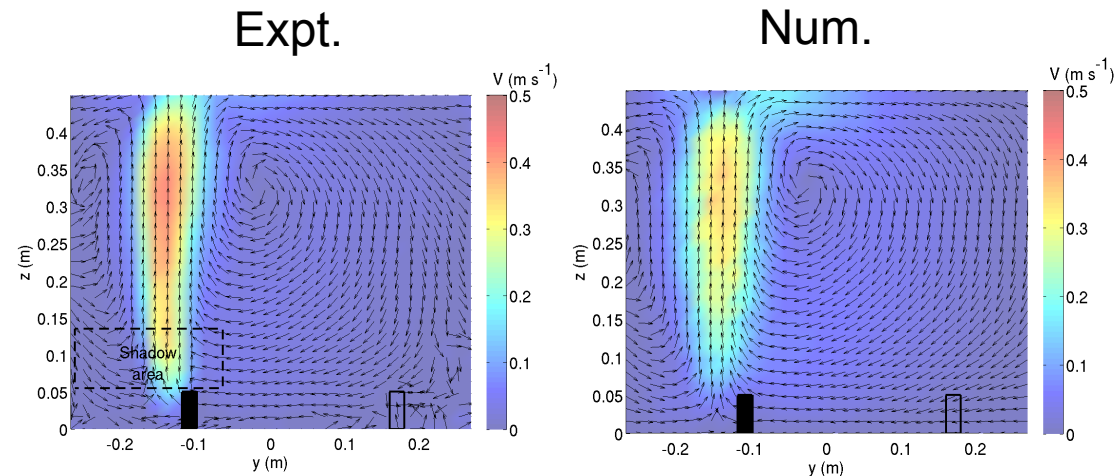
Lagrangian equation
for the trajectory

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

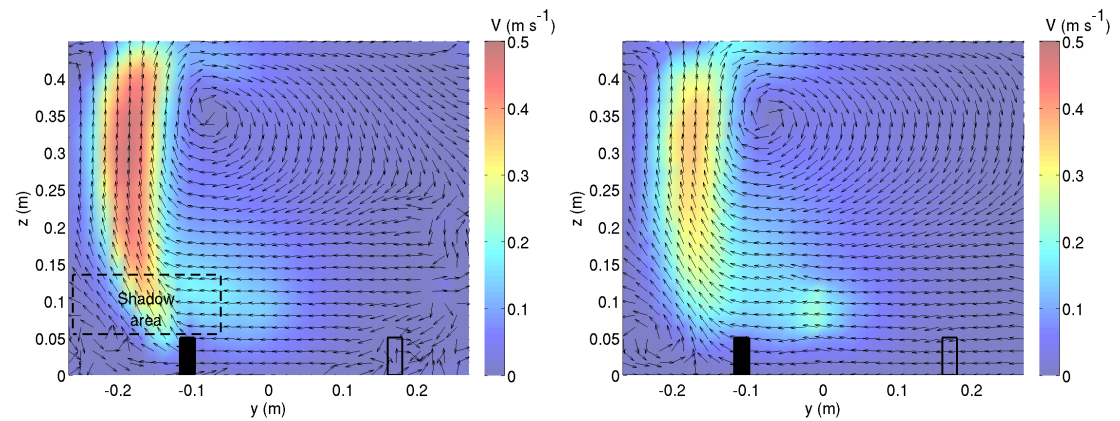


CFD of the vitrification processes : gas bubbling + mechanical

Rotating speed
0 rpm



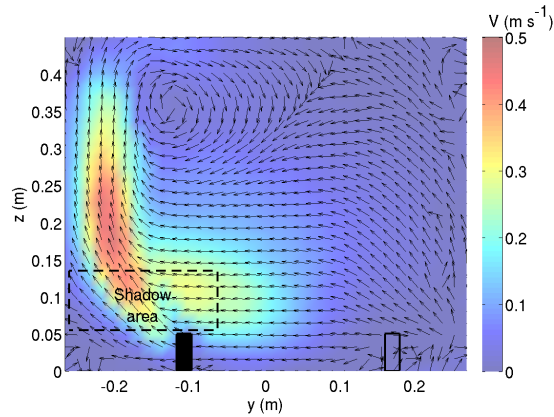
20 rpm



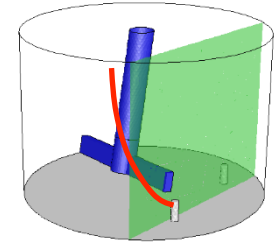
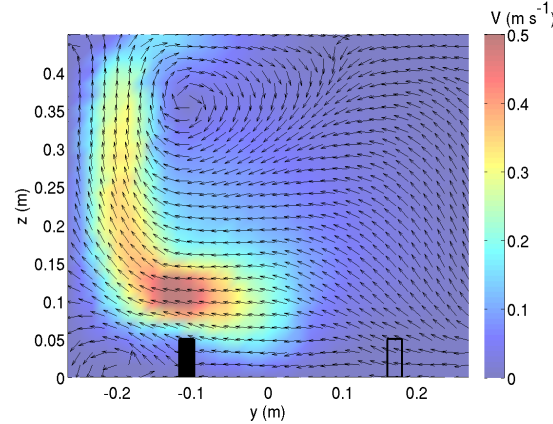
CFD of the vitrification processes : gas bubbling + mechanical

Stirring speed
40 rpm

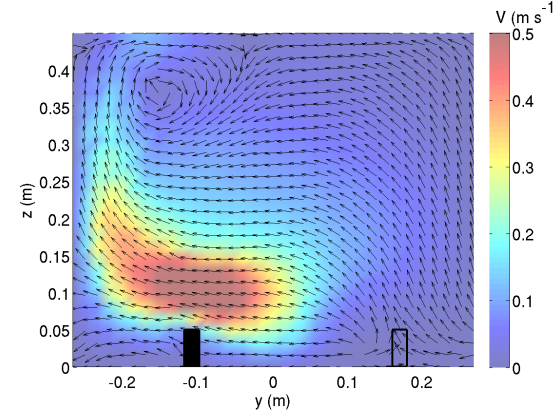
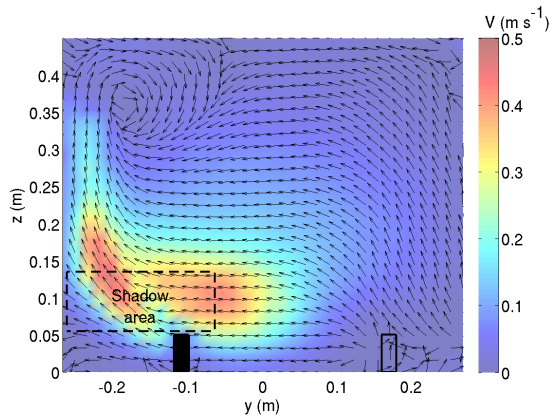
Expt.



Num.

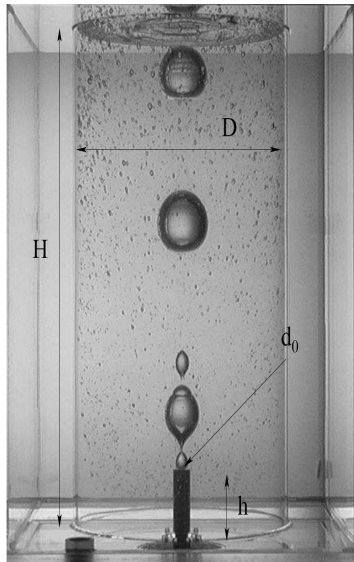


60 rpm

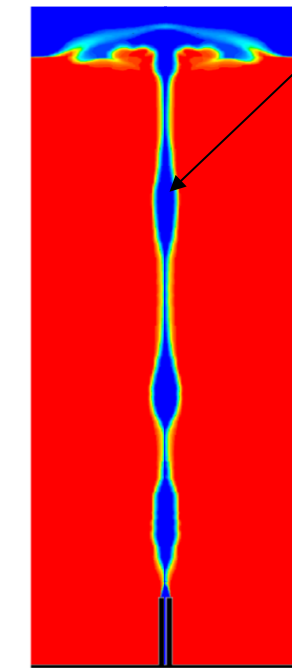


CFD of the vitrification processes : gas bubbling

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

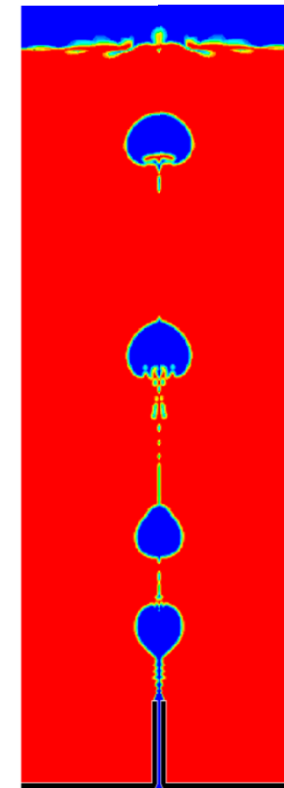
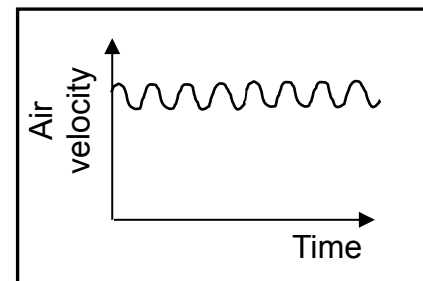


Problem :
No
bubbles
are formed



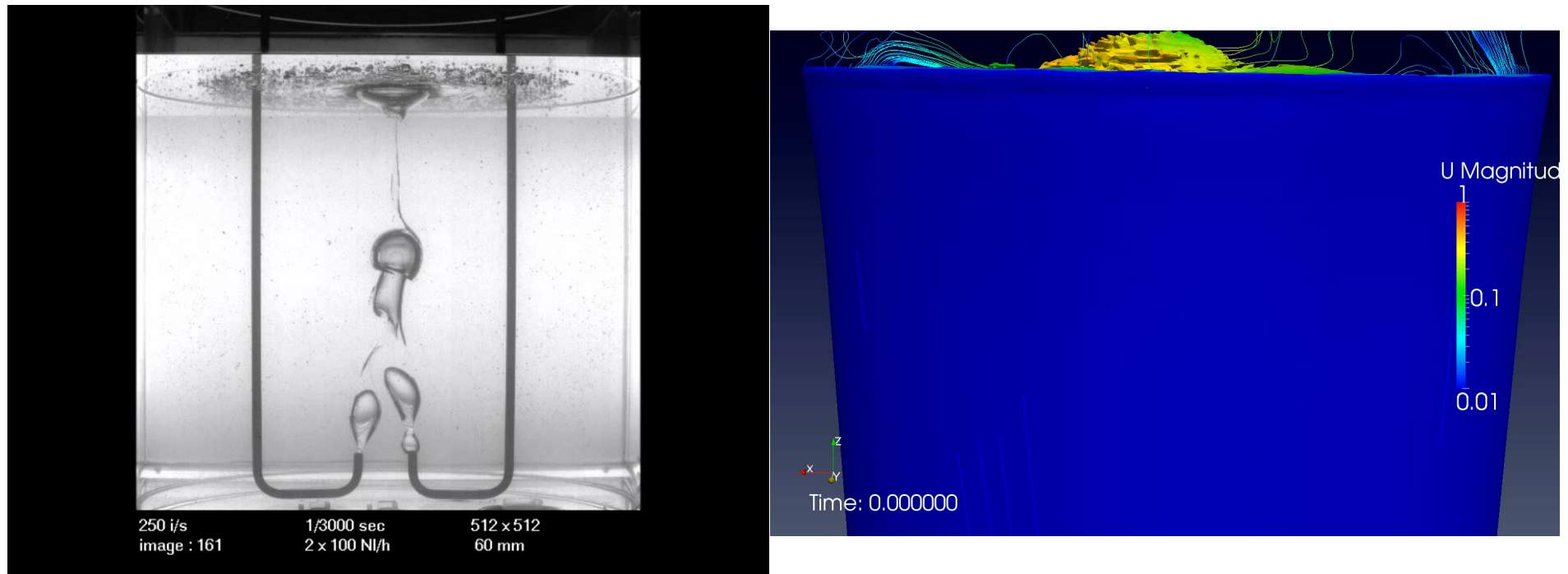
Air jet is a Rayleigh-Plateau instability. This instability should be numerically excited.

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



Complex diphasics flows have been successfully simulated.

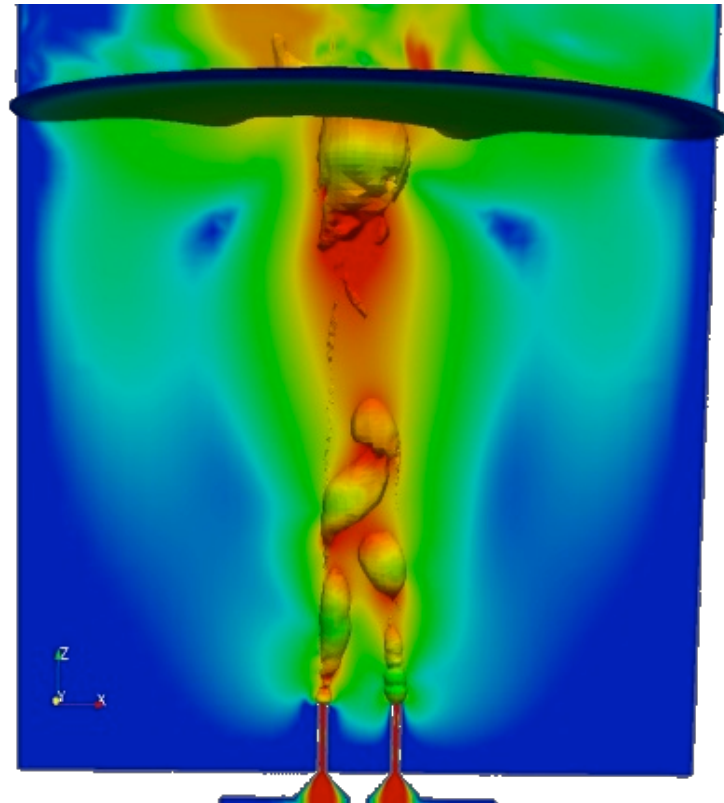
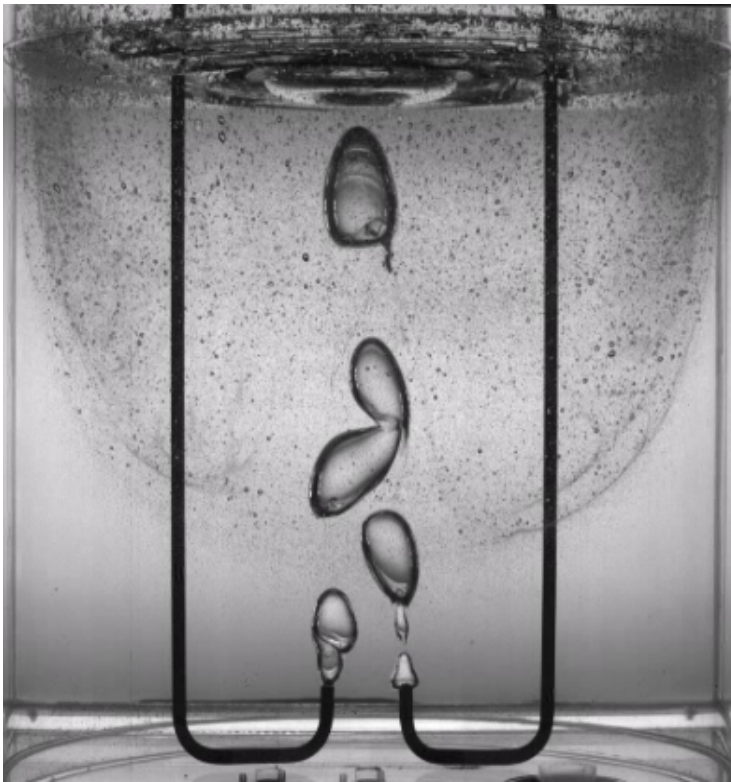
Slow motion (real time / 10)



Main Use :

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

Complex diphasics flows have been successfully simulated.



Main Use :

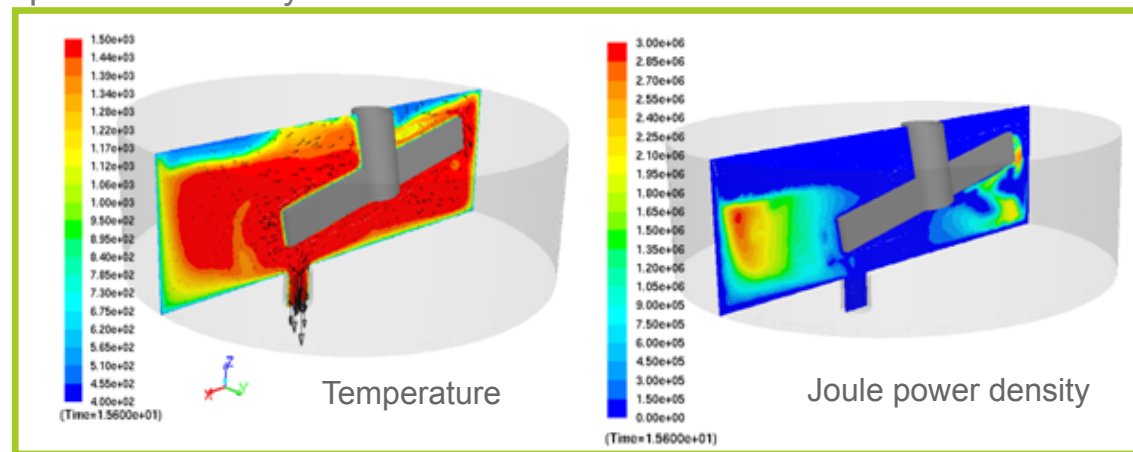
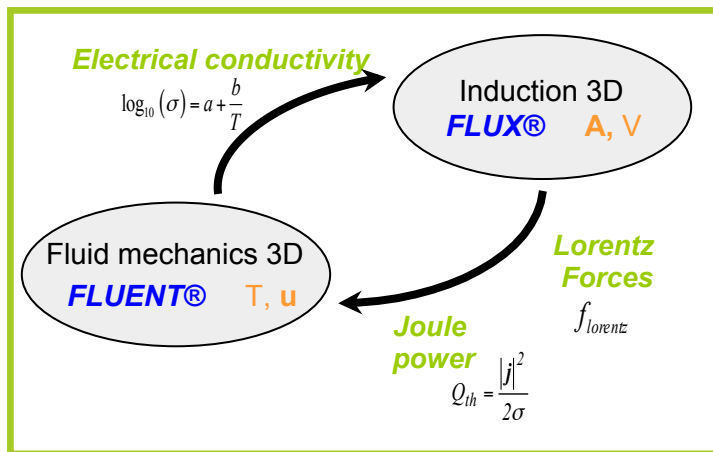
- Study the impact of the bubble formation on the water-cooled bubblers
- Find optimal position of the bubblers to enhance the convection

2 – Coupled phenomena (1/2)

Coupling the physics

- First coupled simulation between induction, CFD and thermal simulation of molten glass with Flux-Expert in 1992. All computations were axisymmetric.
- 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.

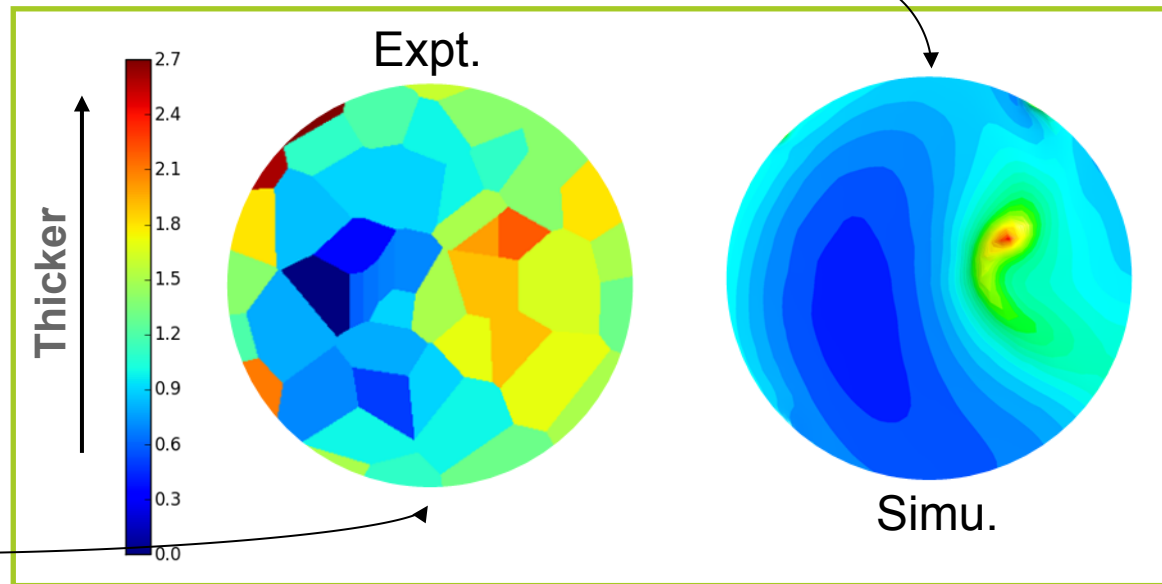
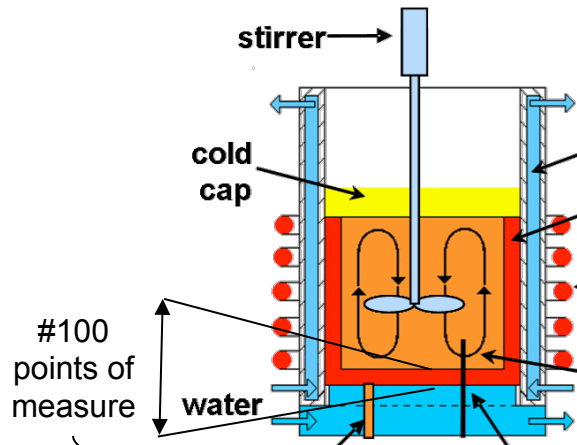
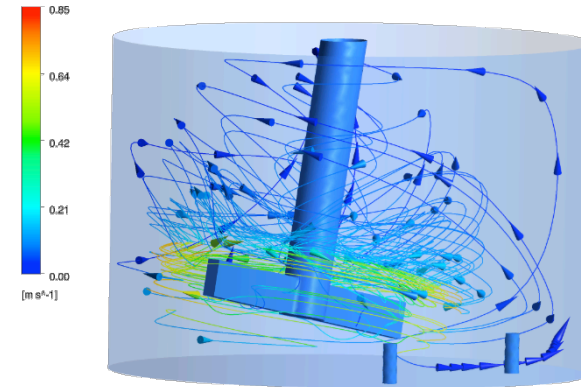
■ Use of specific boundary conditions



2 – Coupled phenomena (2/2)

Validation example : Skull melter thickness at the bottom of the crucible

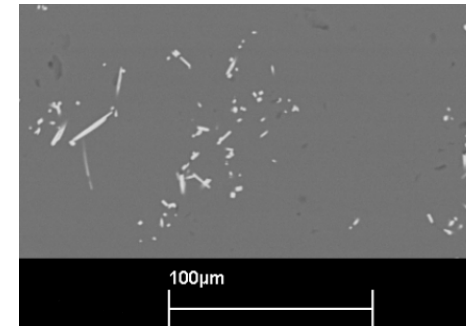
- Inactive glass
- One week long experiment
- Good agreement between simulation and expt. data
- Validation of the fluid flow simulation



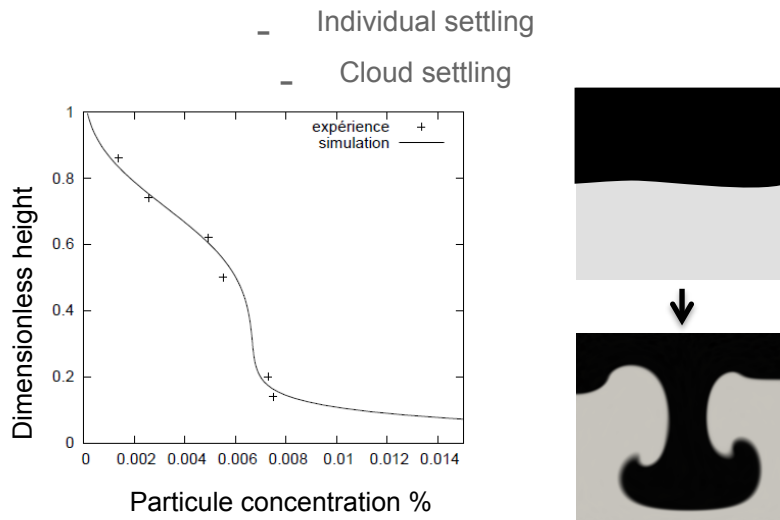
Noble metal particles

- 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 (Pd) spheres
- Non negligible impact on CFD and induction

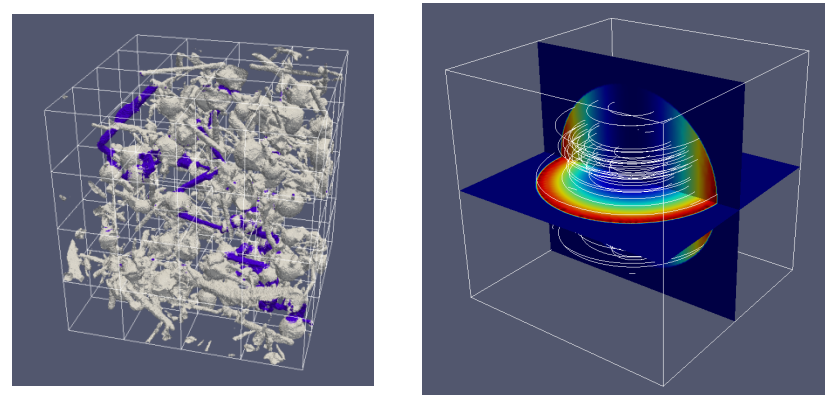
SEM picture of simulated nuclear glass



CFD : Sedimentation phenomena



Induction : Effective electrical conductivity



Noble metal particles settling

■ PHD work G. Barba Rossa

Individual settling (Eulerian model)

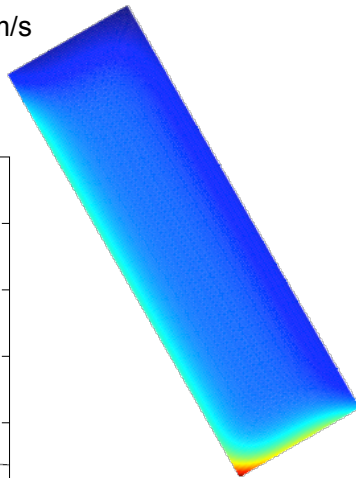
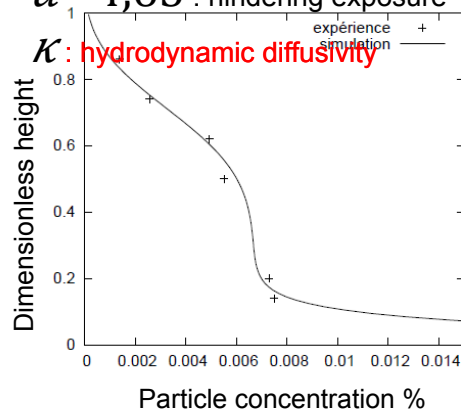
$$\frac{\partial C}{\partial t} + \mathbf{u} \cdot \nabla C + \nabla \cdot (\tau \mathbf{g} C (1 - C/C_{\max})^\alpha) = \nabla \cdot (\kappa \nabla C)$$

τ : Stokes terminal velocity $\sim 1 \mu\text{m/s}$

C_{\max} : maximum concentration

$\alpha = 4,65$: hindering exposure

κ : 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

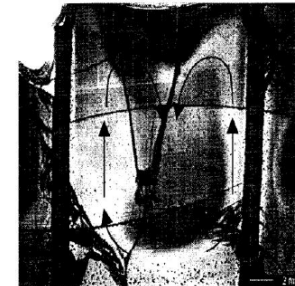
Density of glass depend on the particles concentration :

$$\rho(C) = \rho_f (1 + \beta \downarrow p C)$$

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

$$\beta \downarrow p = \rho \downarrow p / \rho_f - 1 \approx 2,1$$

↓ $s > 0$

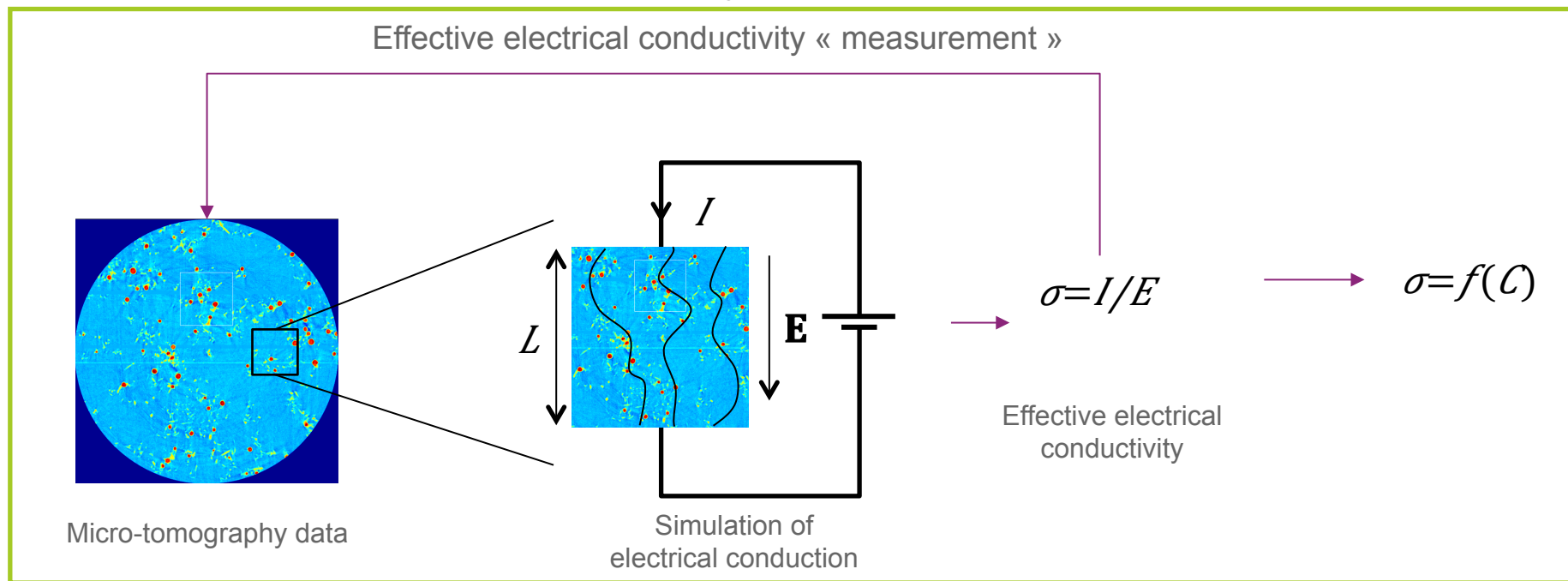
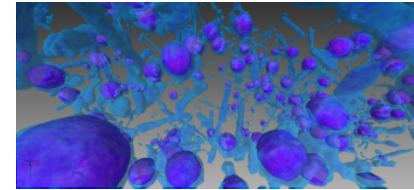


$$s \downarrow \max \uparrow - 1 \propto \eta \uparrow 1/3 (\beta \downarrow p \Delta C) \uparrow - 2/3 \sim 1 \text{ s}$$

Klouzek et al.

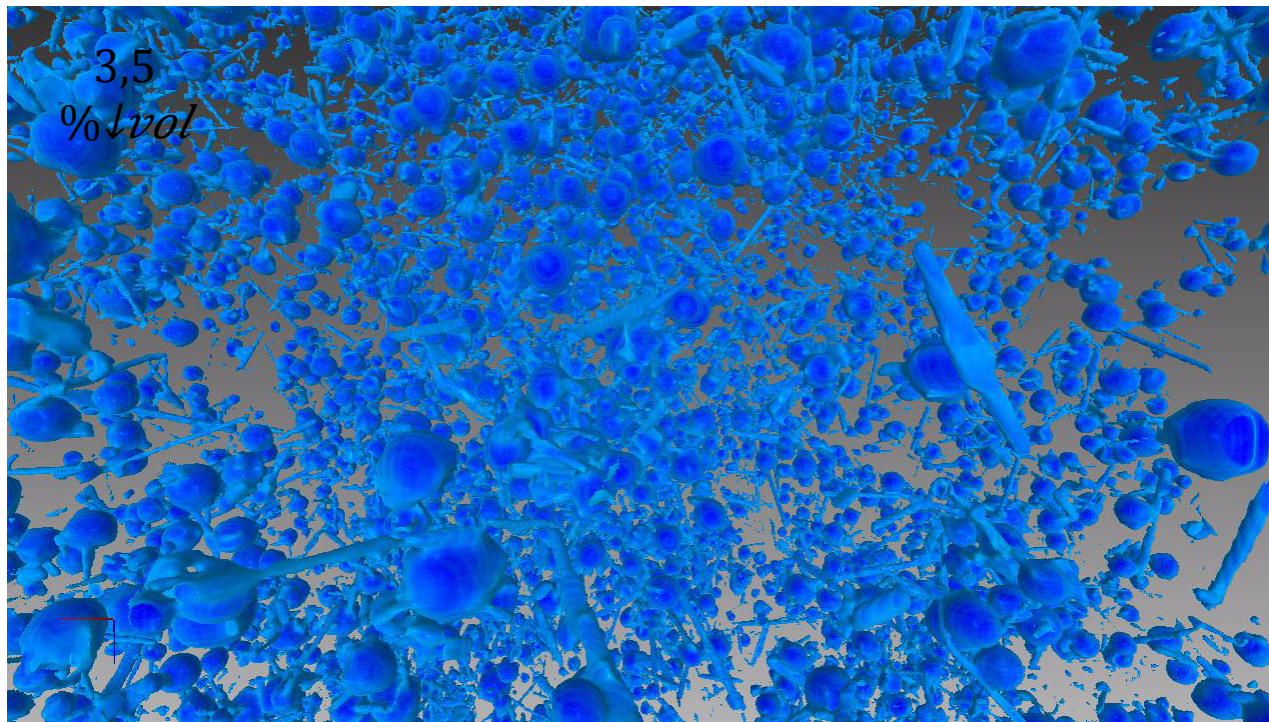
Noble metal particles effect on electrical conductivity

- 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



X Micro-tomography

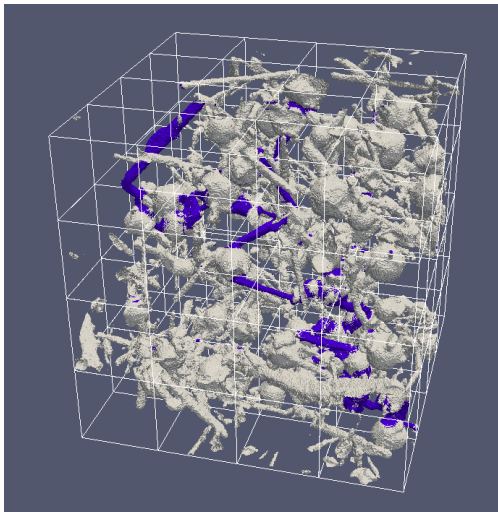
- 3 samples of inactive glass send to the European Synchrotron Radiation Facility (ESRF, ID19)
 - Voxel size of 0,1673 μm



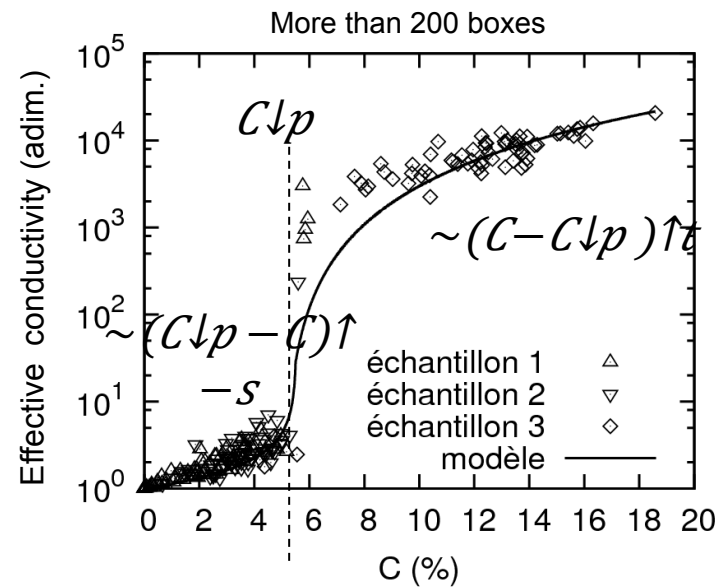
Noble metal particle effect on electrical conductivity

- Micro-tomography X
- Simulation at low scale

Example of a computation box



Percolation curve



Garboczi et al. 1995

$$C \downarrow p \approx 0.6 / \Psi$$



aspect ratio $\Psi \approx 10$

$$\sigma = f(C, C \downarrow p, s, t)$$

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.			
Simulation			

Summary

1 – French radioactive waste vitrification technology presentation

2 - Vitrification simulation

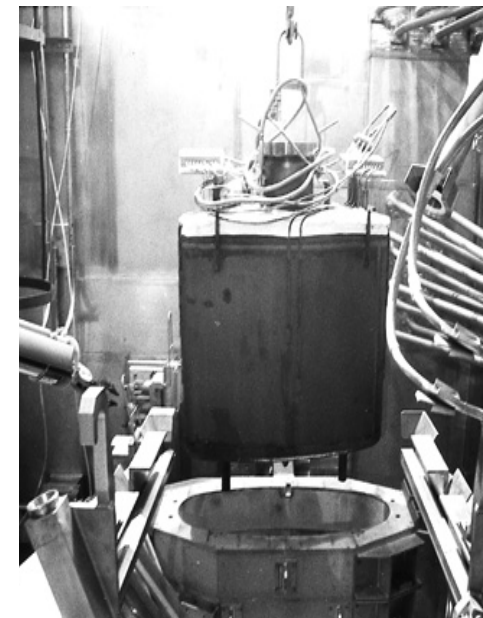
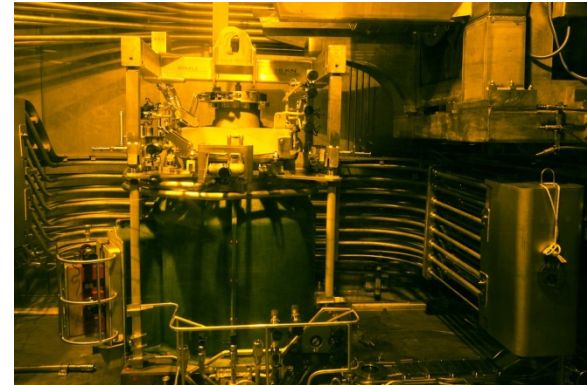
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2-b CFD modelling

2-c Coupled physics

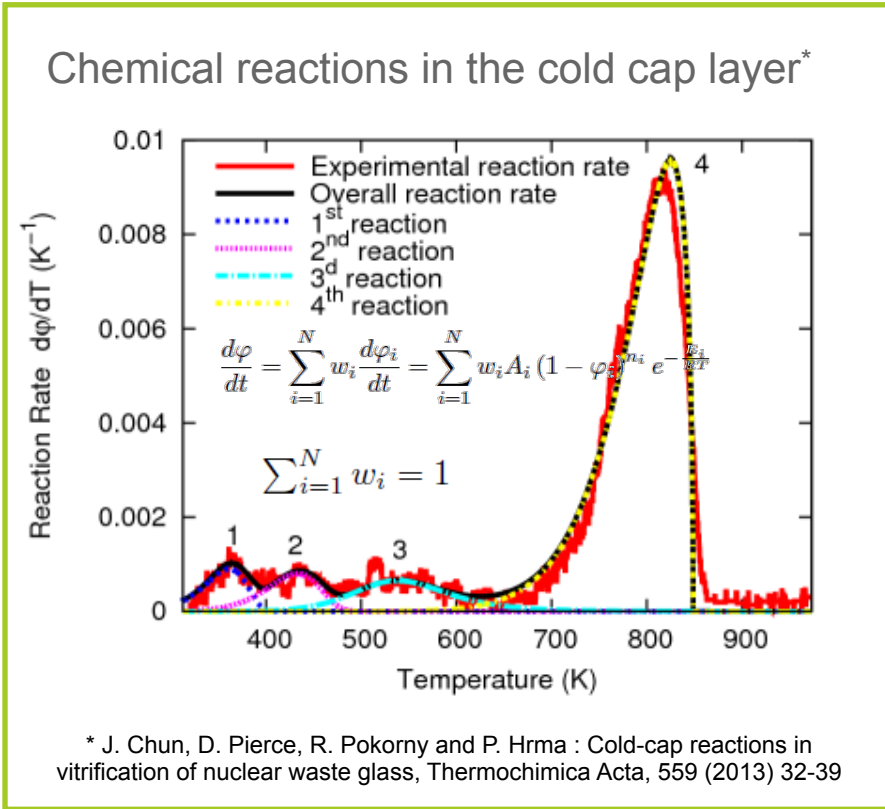
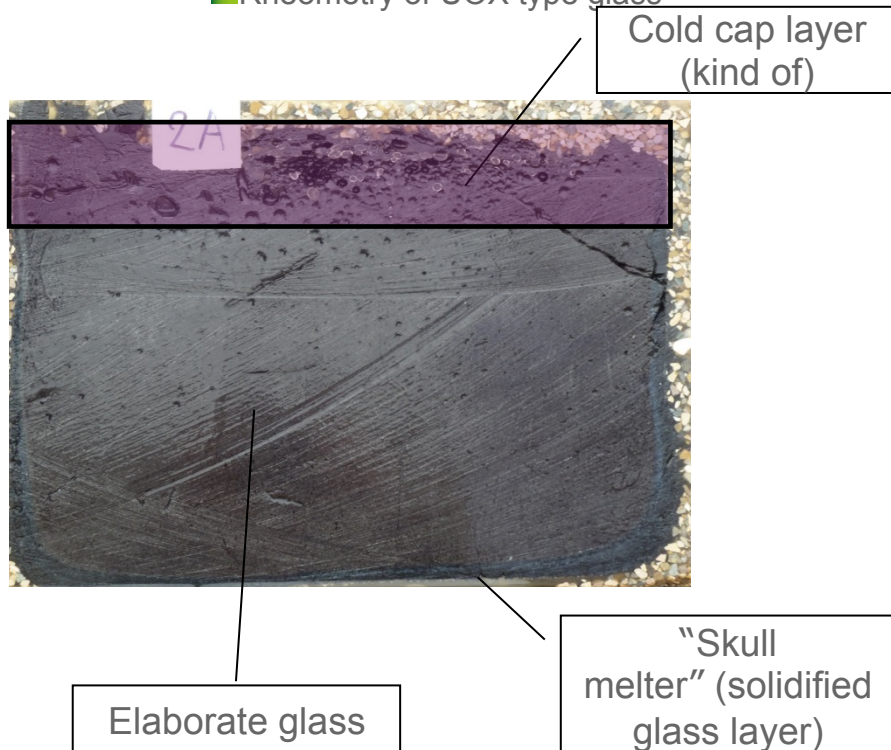
3 – Applications and Ongoing development

4 - Conclusion



Simulation of chemical reactions during vitrification process

- Idea : adapt the work of Hрма – 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 glass¹



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



Thank you for your attention

Any questions ?

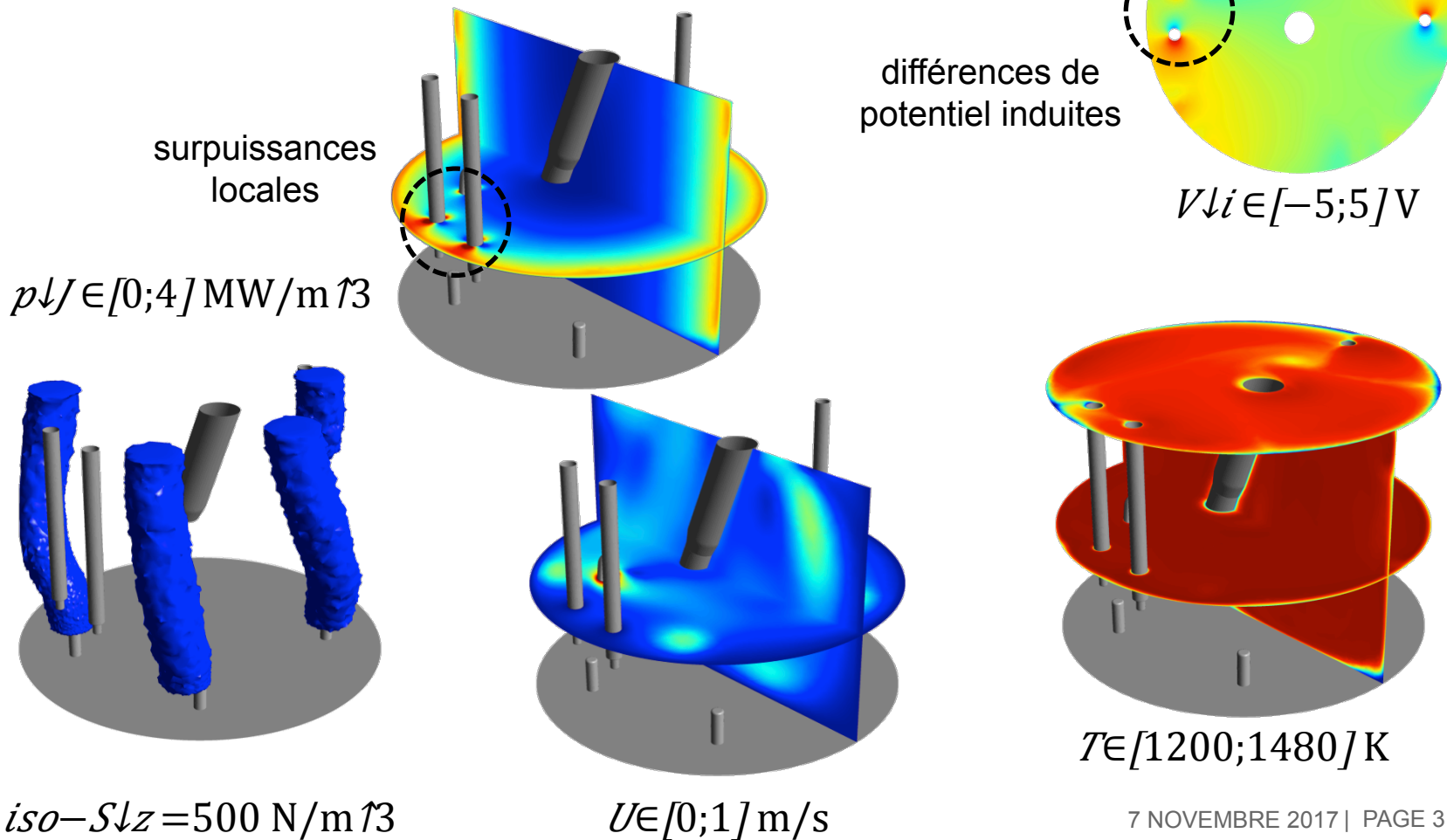
Commissariat à l'énergie atomique et aux énergies alternatives
Centre de Marcoule | 30207 Bagnols-sur-Cèze cedex
T. +33 (0)4 66 79 XX XX | F. +33 (0)4 66 79 XX XX

Direction de l'Energie Nucléaire
DTCD
SCDV

Etablissement public à caractère industriel et commercial | RCS Paris B 775 685 019

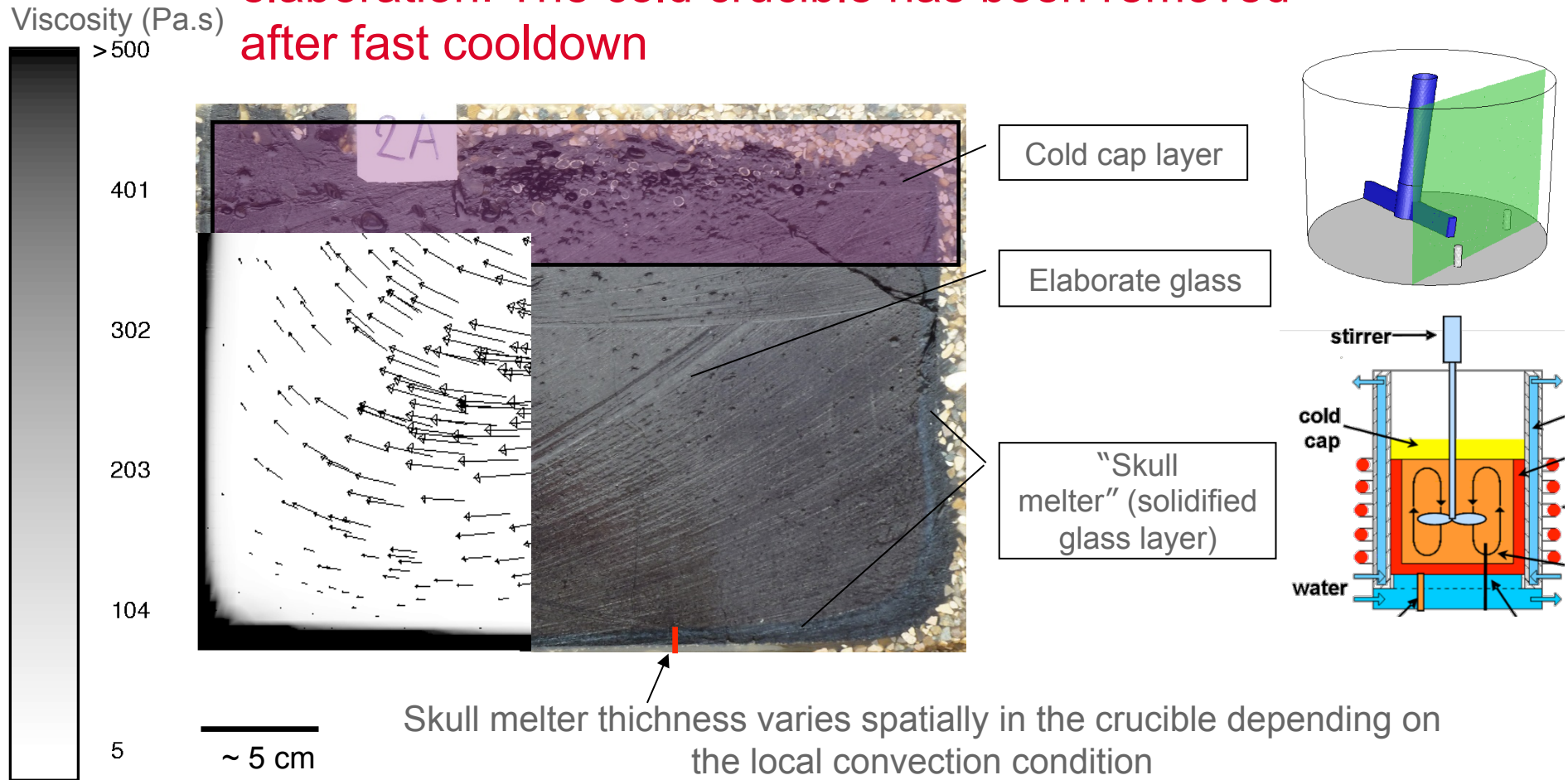
Reponse Magneto-Thermo-Hydraulique

- Champ appliqué $B=1,3 \text{ mT}$, $P \downarrow J = 181 \text{ kW}$
- Température régulée $T \downarrow h = 1473 \text{ K}$



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





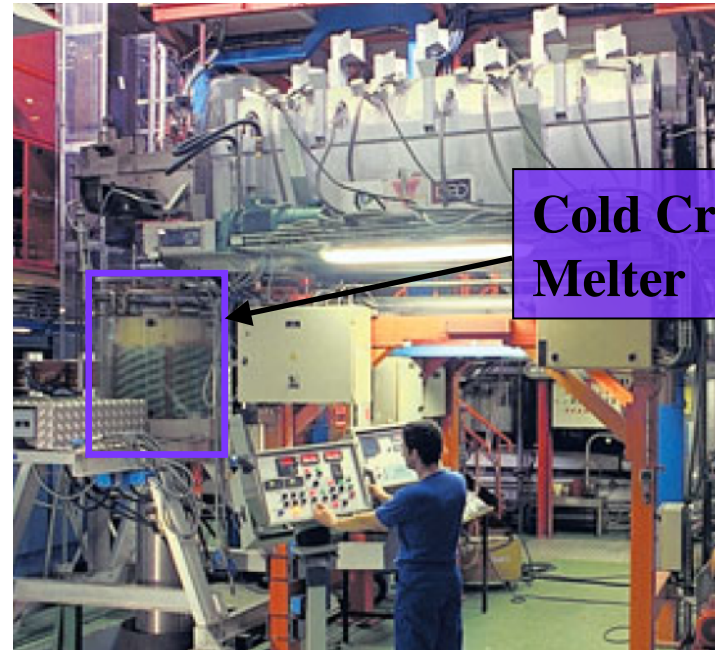
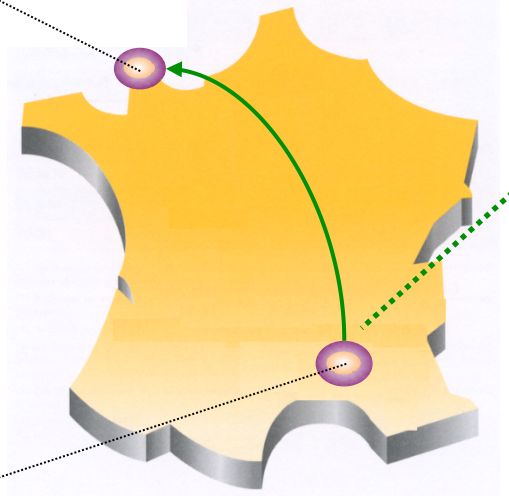
**La Hague
(AREVA)
Industrial plant**



**Marcoule
(CEA)
Research center**

Areva

- Commercial and industrial operator of 2 vitrification plants

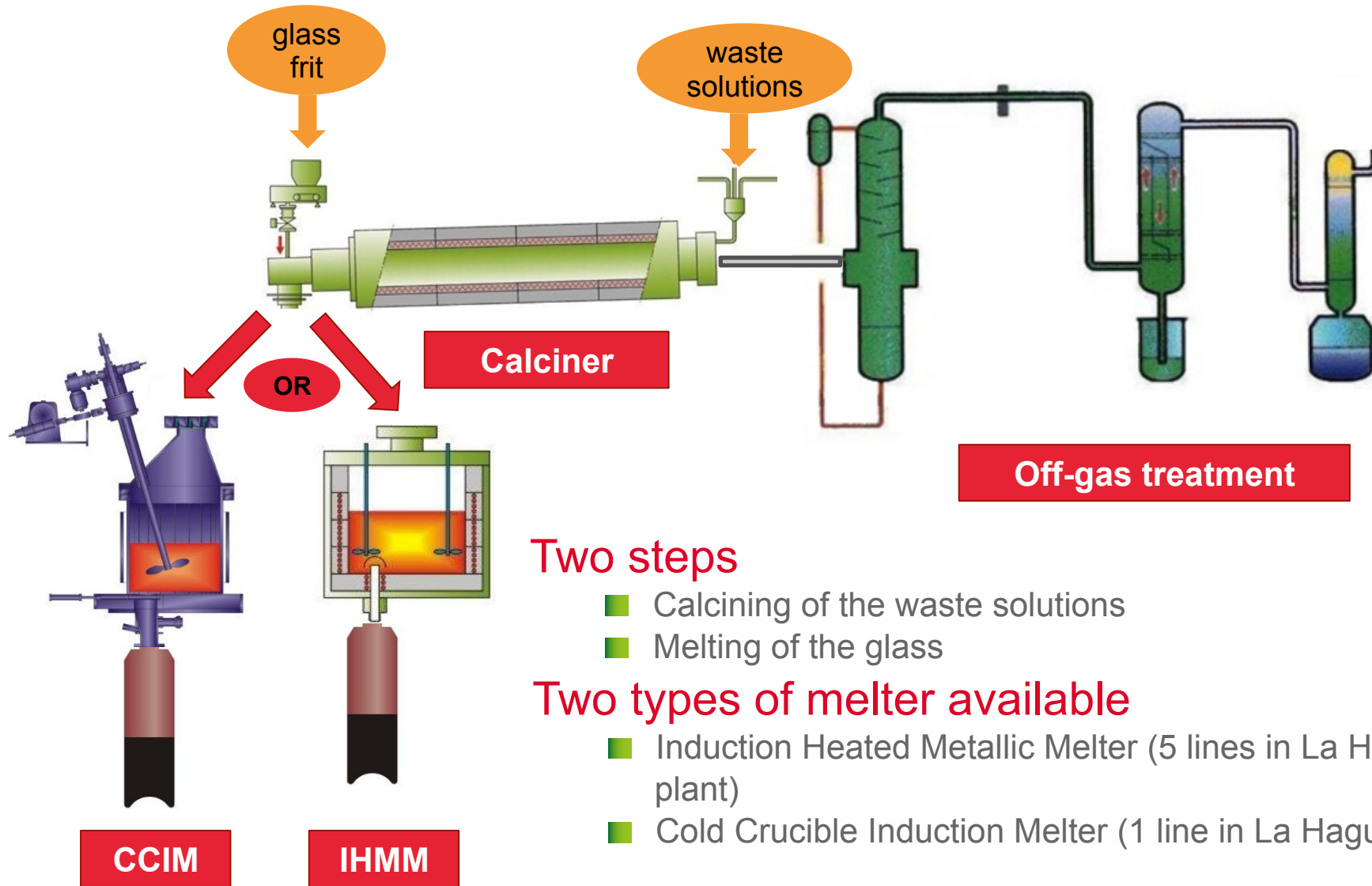


Current pilot of vitrification process
CEA Marcoule (inactive cell)

French Atomic Energy Commission (CEA)

- Confinement Research Engineering Department
Waste Confinement and Vitrification Service

The French vitrification process

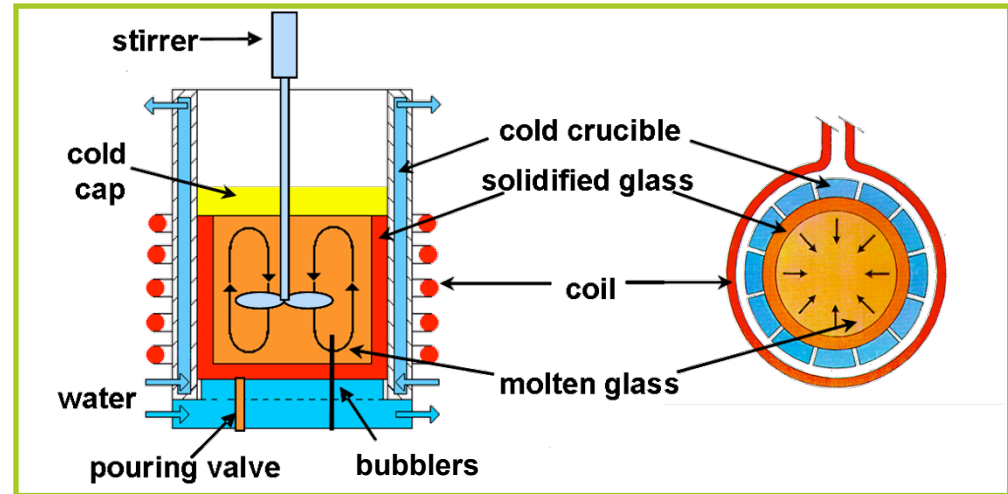


Challenges of the numerical simulation for CCIM

- 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	Unit y	500 K	1500 K
Electrical conductivity	$\Omega^{-1} \cdot m^{-1}$	10^{-5}	20
Dynamic viscosity (μ)	$P a \cdot s$	10^{14}	1
Specific heat (C_p)	$J \cdot kg^{-1} \cdot K^{-1}$	900	1500
Thermal conductivity (λ)	$W \cdot m^{-1} \cdot K^{-1}$	1	6
Density (ρ)	$kg \cdot m^{-3}$	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)