3-D THERMAL, HYDRODYNAMIC & MAGNETIC MODELLING OF ELABORATION OF GLASS BY INDUCTION IN COLD CRUCIBLE

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ABSTRACT

The Vitrification of high-level liquid waste produced from nuclear fuel reprocessing has been carried out industrially for over 30 years by AREVA, with two main objectives: containment of the long lived fission products and reduction of the final volume of waste. In parallel the French Atomic Energy Commission (CEA), AREVA (industrial Operator), and SGN (AREVA's Engineering) have developed the cold crucible melter vitrification technology to obtain greater operating flexibility, increased plant availability and further reduction of secondary waste generated during operations.

The 3D numerical simulation of elaboration of glass by induction in cold crucible needs a coupled approach of the different phenomena: induction, thermal and hydrodynamic. Indeed, those three phenomena are strongly coupled because of the temperature dependence of the glass properties. The hotter the molten glass, the higher the electrical conductivity.

In the present paper, we will focus on a full 3D simulation, when mechanical stirrer and bubbling are stopped in the cold crucible melter. In this case, the convection is driven by two phenomena. First, buoyancy forces are modeled in the Boussinesq approximation. Second, thermocapillary convection at the surface is taken into account. This effect is due to the variation of the surface tension with the temperature. Hydrothermal waves appear at te free surface of the glass bath when the total Joule power injected reached a specific threshold. Qualitative comparison of the aspect of free surface is performed with experimental results.

INTRODUCTION

Vitrification of high-level liquid waste is the internationally recognized standard to both minimize the impact to the environment resulting from waste disposal and the volume of conditioned waste. In France, the vitrification of high-level liquid waste produced from nuclear fuel reprocessing has been successfully operating now for more than 30 years with three major objectives: durable containment of the long-lived fission products, minimization of the final waste volume and operability in an industrial context. As a result, CEA & AREVA have integrated a unique experience in the field of high level waste vitrification through the design and operation of facilities with high records of safety, reliability and product quality, in line with efficient reprocessing plants; continuous efforts to improve at the same time the technology (from hot to cold crucible) and the associated matrix formulations, with constant emphasis on quality and volume reduction, ended up with the design and qualification of the cold crucible melter (CCM) technology.

The cold crucible is a compact water-cooled melter in which the radioactive waste and the glass additives are melted by direct high frequency induction. The cooling of the melter produces a solidified glass layer that protects the melter's inner wall from corrosion. Because the heat is transferred directly to the melt, high operating temperatures can be achieved with no impact on the melter itself. The stirring and air bubblers parameters are optimized to ensure thermal homogeneity while maintaining a cold layer on the surface of the melt. Despite the high temperature of the molten glass (1250°C), this layer limits the quantity of volatilized material.

This technology benefits from the 20 years of french HLW vitrification experience and ensures a virtually unlimited equipment service life and extensive flexibility in dealing with different types of waste. The process and the associated technologies have been also qualified on full-scale prototypes at the CEA pilot facility in Marcoule and at the AREVA pilot facility near La Hague. In parallel, process results are completed by 3D numerical simulation in order to confirm that physical phenomena are well controlled.

Several publications [1,2] have been already presented on the description and advantages of the CCM for various applications and matrix design. In the present paper, we will focus on the 3-D thermal, hydrodynamic & magnetic modelling of elaboration of glass by induction in cold crucible.

PROCESS AND TECHNOLOGY

The cold crucible melter was substituted in the process for the existing metal pot beneath the calciner. It was designed to meet environmental and process constraints: for example, the canister is still filled by pouring successive melts.
The use of the CCM has no effect on the other process equipment (calciner and off-gas treatment); the overall performance of the process line complies with the initial design rating of the vitrification line.
The choice of conserving a 2-step process (separation of the calcining and melting function, **Fig. 1**) was justified by compliance with the basic concept implemented by AREVA for all its equipment (compact, modular design, allowing easy maintenance and resulting in a small volume of technological and secondary waste) and by the objective of minimizing the complications and investment cost of incorporating the unit in the existing line.

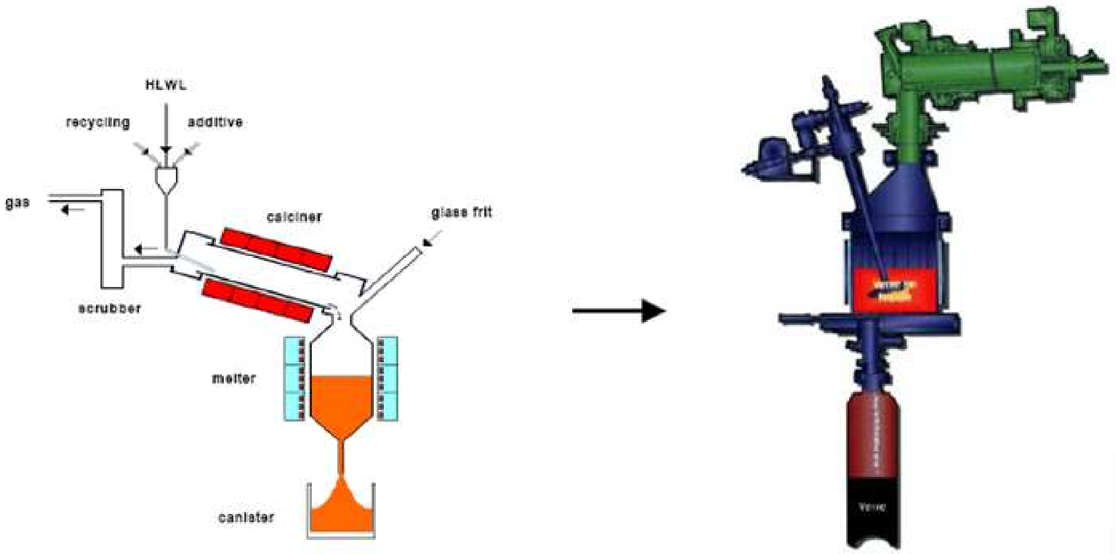


Fig. 1. Two-step vitrification process

The melter design complies with all remote maintenance principles applicable in AREVA vitrification cells; it is a compact one-piece unit to simplify installation and removal. Its light weight (less than 1 ton) and 'cold' design ensure optimization of the ultimate technological and secondary waste volume and suitability for incorporation in the existing waste treatment processes at La Hague.
The CCM is a sectorized crucible which is cooled by an internally circulation of pressurized water. The inner diameter of the cold crucible melter is 650 mm.
The load is heated by direct induction thanks to the moderate electrical conductivity value of the glass at high temperature. A high frequency generator supplies the power necessary to fabricate the glass. The inductor is outside and connected to a high frequency generator.

The molten glass temperature is an important parameter for ensuring product quality, particularly by maintaining a minimum fabrication temperature to guarantee a suitable vitreous state. This is why, a mechanical stirring and air bubblers are added: natural convection and Laplace forces are too weak compared to its high viscosity to assure a good mixing of the melt.
The stirring parameters (rotation speed and direction,…) and air bubblers parameters (flow rate,…) were optimized to ensure thermal homogeneity while maintaining a cold layer on the surface of the melt to limit quantity of volatilized material.

Induction technology development on a full-scale prototype is conducted in parallel with glass formulation studies, and it is always the result of a combined approach which involves close links between research, modelling, engineering and operating team, and also a judicious build up of results and experience.

PURPOSE AND DIFFICULTIES OF NUMERICAL SIMULATION

Numerical simulation first objective is to help to understand physical phenomena within the molten glass. In fact, due to high temperature and high corrosive properties of the melt, accurate experimental data are very difficult to obtain. Thereafter, the simulation can help to optimize the design of structures or the high frequency power line distribution. But also, the simulation can give an evaluation of thermal and chemical homogeneity and the impact of the glass properties on this homogeneity.

The 3D numerical simulation of elaboration of glass by induction in cold crucible needs a complex coupled approach of the different phenomena:

- All the properties of the glass are strongly dependant of the temperature,
- many physics are involved; a thermal equilibrium between volumic heating within the glass and cooling of the wall is present,
- induction, forced and natural convection, radiation take a part in this equilibrium.
- the complex design of the process especially of the CCM and of the mechanical stirrer leads to huge and complex meshes,

PHYSICAL PROPERTIES OF THE GLASS

The glass is supposed to be a Newtonian fluid but all the physical properties are complex functions of the temperature. These laws are confidential but Table I summarizes order of magnitude for two temperatures. The confinement glass of this study is opaque so there is no need of internal radiation model. In fact, the internal radiation is naturally included in the variation of thermal conductivity with temperature.

Table I: Order of magnitude of physical properties of the glass

Physical properties	Unity	500 K	1500 K
Electrical conductivity	$\Omega^{-1}.m^{-1}$	10^{-4}	20
Dynamic viscosity	*Pa.s*	10^{14}	1
Specific heat	$J.kg^{-1}.K^{-1}$	900	1500
Thermal conductivity	$W.m^{-1}.K^{-1}$	1	6
Density	$kg.m^{-3}$	2850	2750

MODELLING OF INDUCTION HEATING COUPLED TO THERMO-HYDRODYNAMIC

This study is in continuation of previous works modeling this process (Jacoutot [3] and Sauvage [5]). The aim is to model the molten glass heated by direct induction. Thus, hydrodynamic, thermal and induction phenomena are taken into account. As the physical properties of the glass are functions of the temperature, these three phenomena are strongly coupled. Consequently, a coupling between two softwares is achieved. Fluent® is used to solve hydrodynamic and thermal equations whereas Flux® computes the induction equations. All computations are three-dimensional. The coupling is based on files data transfers. Each software interpolates the field on the computing nodes of the other software. This full 3D-3D coupling has been presented in previous paper [5]. The coupling is used in another configuration in which for simplicity the mechanical stirring and bubbling is not modeled. Only thermoconvectives forces will acts on the flow. Besides, we will focus on the thermal boundary conditions type.

Geometry

The molten glass bath is a cylinder of 450mm high and 325mm radius. In this study the mechanical stirrer is not modeled, only two thermocouples are taken into account.

Electromagnetic model

The commercial software Flux® is used to solve induction equations. The crucible as well the inductor are not modeled, they are approximated by a current sheet surrounding the glass. In this coil, an alternative current (I_{eff} = 1500 A and 280 kHz) is imposed. Owing to the high value of the frequency, the quasi-steady approximation is made. Different formulations for induction equations are available. The **A**-V formulation gives best result in a material with high gradient of electrical conductivity.

$$\begin{aligned} &\vec{\nabla}\times\left(\frac{1}{\mu_0}\vec{\nabla}\times\left(\vec{A}\right)\right)+\sigma\left(\frac{\partial\vec{A}}{\partial t}+\vec{\nabla}V\right)=\vec{0} \\ &\vec{\nabla}\cdot\left(\sigma\left(\frac{\partial\vec{A}}{\partial t}+\vec{\nabla}V\right)\right)=\vec{0} \end{aligned} \quad (1)$$

The coil is not meshed so a reduced scalar potential formulation is used. The resolution is achieved with iterative methods such as a conjugate gradient one. The domain is discretised with approximately 200 000 first-order elements.

Thermal-Hydrodynamic model

Fluent® software solves the Navier-Stokes and thermal equations. The flow is assumed to be laminar due to the high viscosity of the glass.

$$\vec{\nabla} \cdot \vec{u} = 0$$

$$\rho_0 \left(\frac{\partial \vec{u}}{\partial t} + \left(\vec{u} \cdot \vec{\nabla} \right) \vec{u} \right) = -\vec{\nabla} p + \vec{\nabla} \bar{\bar{\tau}} - \rho_0 \beta \left(T - T_0 \right) \vec{g} \qquad (2)$$

$$\bar{\bar{\tau}} = \vec{\nabla} \cdot \left(\mu \left(\vec{\nabla} \cdot \vec{u} + \vec{\nabla} \cdot \vec{u}^T \right) \right) \qquad (3)$$

$$\rho_0 \left(\frac{\partial c_P T}{\partial t} + \left(\vec{u} \cdot \vec{\nabla} \right) c_P T \right) = -\vec{\nabla} \cdot \left(\lambda \vec{\nabla} T \right) + Q_{th} \qquad (4)$$

The source terms in the right hand side of equation (4) is the Joule power density dissipated in the glass. This term is calculated by Flux® as a function of induced current:

$$Q_{th} = j^2 / 2\sigma \quad and \quad \vec{j} = -\sigma \left(\frac{\partial \vec{A}}{\partial t} + \overrightarrow{grad} V \right) \qquad (5)$$

The convection is driven by two phenomena. First, buoyancy forces are modeled in the Boussinesq approximation (last term of right hand side of equation (2)). Second, thermocapillary convection at the surface is taken into account. This effect is due to the variation of the surface tension with the temperature. This dependence is well described with the law: $\sigma_s(T) = \sigma_{s0} - \gamma (T - T_0)$ where $\gamma = -\partial \sigma_s / \partial T$. For almost all liquid, γ is constant and positive and for the glass its value is $10^{-4}\ N.m^{-1}.K^{-1}$ [4]. The boundary condition at the free surface relates the viscous strain with the thermal strain in the direction of the surface, for example, in the radial direction:

$$\mu \left. \frac{\partial u}{\partial z} \right|_{surf} = \gamma \left. \frac{\partial T}{\partial r} \right|_{surf} \qquad (6)$$

Thermal boundary conditions choice for the cooled parts of the process is discussed in the next section. At the free surface a mixed condition convection-radiation is considered with an emissivity of 0.9 and a heat exchange coefficient of 20 $W.m^{-2}.K^{-1}$.

Thermal boundary conditions

The skull melter which is solidified glass in contact with the cooled wall of the process, is very important for the final quality of the glass. High thermal gradient takes place in it leading to specific difficulties during numerical simulation. In consequence, specific investigation is going to be performed on the thermal boundary condition of the glass. Two different types of condition will be tested:

1- Fourier condition: thermal exchange coefficient h ($W.m^{-2}.K^{-1}$) and bulk temperature T_∞ (K) are imposed:

2- Dirichlet condition: specific temperature value is imposed

For the first type, heat transfer coefficients are supposed to be uniform on a given cooled part of the process, like sector, bottom or thermocouple rod. "h" depends of the internal geometry of the element. One dimensional approach is used in order to evaluate them. The glass is supposed to be in contact of the inox wall of thickness e_{inox} (m) which is cooled by an internal circulation of cold water. Then expression of the equivalent global exchange coefficient is:

$$h = \frac{1}{\frac{e_{inox}}{\lambda_{inox}} + \frac{1}{h_{convection}}}$$

with $h_{convection}$ ($W.m^{-2}.K^{-1}$) representing the thermal exchange coefficient in the water boundary layer. Experimental correlation is available in the literature to estimate it:

$$Nu = \frac{h_{convection} D_h}{\lambda_{water}} = 0.00243\,\mathrm{Re}^{0.8}\ \mathrm{Pr}^{0.4}$$

with D_h (m) the equivalent diameter of the water flow section, Pr the Prandtl number of the water, Re the Reynolds number calculated with the average velocity of water and λ_{water} the thermal conductivity of water. Previous equations give different results function of the thickness of inox and velocity of cooled water. The results in our configuration and standard operating conditions are 3000 for the thermocouple tubes and 1000 for the sector of the crucible. These value will be used as thermal boundary condition with a bulk temperature of the water of T_∞=350 K.

Specific difficulties are related to this kind of boundary condition. First of them is the determination of the coefficients characterizing the internal exchange efficiency when the mono-dimensional approach is no longer valid. The second difficulty was point out by Jacoutot [2] which has shown that values of these coefficient estimated as previous give in some case non-realistic results: temperature calculated at the wall were too high in comparison with experimental measurement. These overheat are located where thermal flux is very high corresponding to the thinner skull-melter zone. The reason is probably the huge thermal gradient in these zones (up to 5.10^4 $K.m^{-1}$) which is very difficult to catch with numerical simulation because of numerical diffusion.

Therefore, another thermal boundary condition is tested: Dirichlet condition which means to impose the value of the temperature. We consider that isotherm 800 K is the limit of the skull-melter layer because for this value of temperature glass has a viscosity of 10^4 Pa s. Experimental observations give us an average thickness for the skull-melter of 5 mm. This thickness is neglected in front of the crucible diameter of 650mm and a temperature of 800 K is directly imposed at the wall.
The main advantage of this condition is that no coefficients have to be determined. Another advantage is that thermal gradient is diminished which reduce numerical diffusion and increase the resolution speed. As the cold glass is solid, adherence boundary condition is still valid for hydrodynamic calculations.
The two type of thermal boundary conditions are going to be used and compared in the academic configuration.

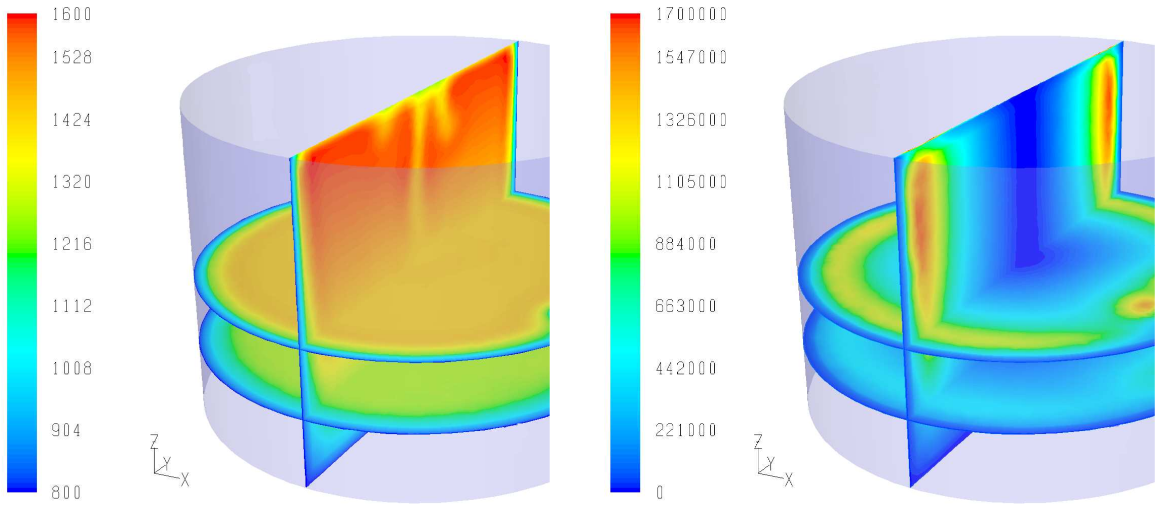
Coupling Strategy

The main issue is to couple a finite element (Flux®) and a volume finite (Fluent®) based software. Mesh refinement requirements are different for the induction and hydrodynamic phenomena. Consequently, using a unique mesh is not possible and interpolations between the two meshes are done to minimize losses of precision. These methods have to be robust and fast in order to not penalize computation time. Each software interpolates the field on the computing nodes of the other software. Flux® software natively includes these methods but specific functions have been created to allow Fluent® to do these interpolations using the calculated temperature gradient in each cell.

The Joule power distribution is calculated every 10s second of flow computation. This time was chosen in regard of the characteristic convection time which is about 500*s*. As Jacoutot [3] shown, this does not change the results. The flow time step for Fluent® is 1s.
The convergence criterion for the iterative coupling is based on the square error between the old and new map of temperature. It is calculated by the software Flux® during importation of the new field of temperature, the convergence threshold is set at 10^{-5}.

RESULTS for a fixed total Joule power of 80 kW

A calculation is performed with the 3D iterative coupling of an academic configuration without stirring systems.



(a) (b)

Fig. 2. Temperature (K) isocontours (a) and Joule power density (W m^{-3}) isocontours (b) on crossing planes. Figures are cut for confidentiality reasons.

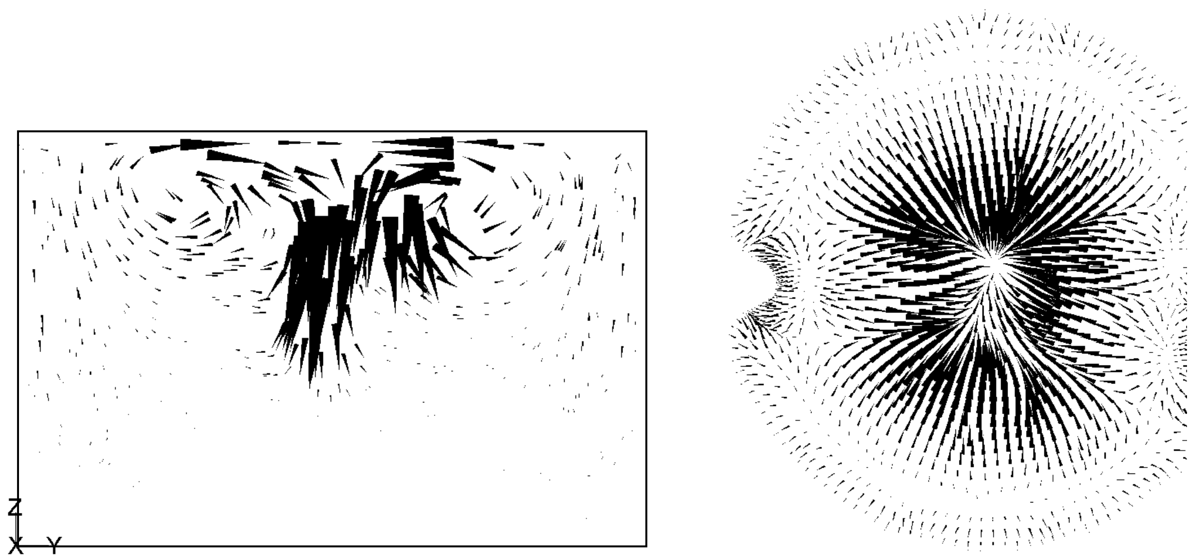
Figure 2 shows the temperature field and the corresponding Joule heat sources calculated with Flux. The temperature filed exhibits the presence of the skull-melter along cooled walls. The flow is quite stratified because thermoconvective forces are not strong enough to assure a good thermal homogeneity. That is why, there is in real process a mechanical stirrer and air bubbling stem.

Table II. Global heat flux repartition in the two studied configuration

		Heat flux (kW)		
Boundary type	T_{max} (K)	Crucible	Free surface	Bottom
h and T_∞	1623	38.1	28.3	4.9
T_0 = 800K	1615	38.9	29.1	3.3

Table II present thermal flux repartition for a fixed total Joule power injected of 80 kW found with the two type of boundary condition. Results obtained with the two type of boundary are really similar. Temperature and repartition of thermal flux are very close. The main difference is for the bottom which can be explained by the fact that this part has the greatest skull-melter thickness.
In a stirred case, glass temperature is totally homogenous and skull-melter thickness is much thinner [3]. Consequently the method of imposing a temperature will be appropriate.

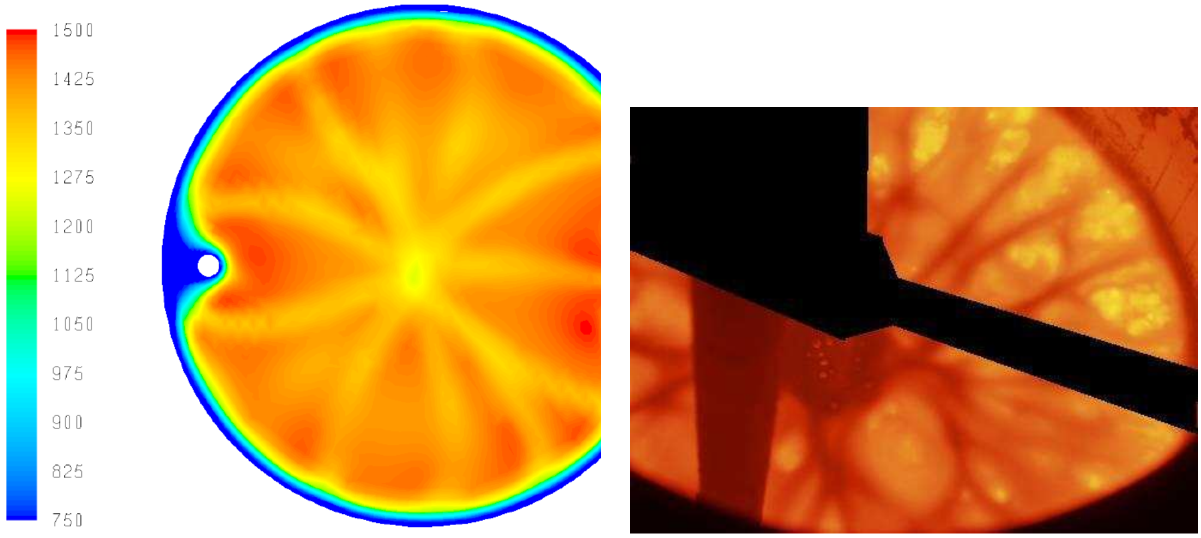
Flow configuration



(a) (b)

Fig. 3. Velocity vectors (m s^{-1}) on (a) a vertical plane (V_{max}=6 mm.s^{-1}) and (b) at the free surface of the melt (V_{max}=4 mm.s^{-1}).

The figure 3-a shows the velocity vectors on a vertical crossing plane. The two classical recirculation cells are visible.
For this amount of Joule power injected in the glass, the flow shows hydrothermal waves at the free surface due to Marangoni convection (figure 3-b and 4-a). Note that no vertical deformation of the free surface is taken into account. These waves are thermal waves propagating mainly in the azimuthal direction in the pool (see [6] for an academic study of such waves in oil flow). A qualitative comparison between simulated and experimental view of these waves at the free surface is shown on figure 4-b.



(a) (b)

Fig. 4. (a) Temperature isocontours (K) at the free surface showing hydrothermal waves, and (b) experimental view of similar flow configuration (CEA Marcoule).

CONCLUSION

The 3D coupling between thermo-hydrodynamic and induction phenomenon in a glass melt previously developed [5] has been successfully applied to a new process configuration including intrusive cooled metallic elements in the glass melt. Two types of thermal boundary conditions have been tested. The method of imposed temperature shows advantages like to have no coefficient to determined, less numerical diffusion and better convergence compared to Fourier condition. Its main assumption which is to neglect the skull-melter thickness has been proven to have low impact on final results.

Moreover, if a certain threshold of the total injected power is reached, then 3D hydrothermal waves appear at the free surface due to Marangoni convection. This pattern is time-dependent and presents chaotic behavior. Qualitative comparison has been performed with experimental observation. It should be specified that this flow pattern is found in this example only because mechanical stirring and air bubbling are not modeled. In nominal conditions of the process, these movements are totally erased by forced convection. But the capability of the coupling method to simulate such complex flow configuration is a good proof for validation.

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