3-D Thermal, Hydrodynamic & Magnetic Modelling of Elaboration of Glass by Induction in Cold Crucible - 9033

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ABSTRACT

The Vitrification of high-level liquid waste produced from nuclear fuel reprocessing has been carried out industrially for more than 30 years by AREVA, with three main objectives: containment of the long lived fission products, reduction of the final volume of waste and operability in an industrial context. In parallel the French Atomic Energy Commission (CEA), SGN (respectively Areva's R&D provider and Engineering) and AREVA (industrial Operator) have developed the cold crucible induction 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 modelled in the Boussinesq approximation. Second, thermo capillary convection at the surface is taken into account. This effect is due to the variation of the surface tension with the temperature. Thermo convectives circulations appear within the molten glass when the total Joule power injected reached a specific threshold.

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 and SGN 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.

CCM technology has brought the following improvements:

- A significantly extended lifetime and thus minimisation of maintenance operations and secondary waste,
- An increased flexibility towards waste and glass compositions owing to its corrosion resistance and the ability to reach high temperatures. Thus, it allows developing higher performance matrices accessing to greater amount of refractory material (Zr,Al, Si) or to compositions with higher waste loading factors. It also allows the fabrication of glass ceramics (Mo, Zr, F) or ceramics (Synroc, Zirconolite).
- A relief in the operating constraints as limiting the settling of undissolved elements by the use of a mechanical stirring that also improves the product quality (homogeneity).

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 a full-scale prototype at the CEA pilot facility in Marcoule. 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.

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 incorporating the unit in the existing line.

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 400 kW generator operating at a frequency of 300 kHz supplies the power necessary to fabricate the glass.

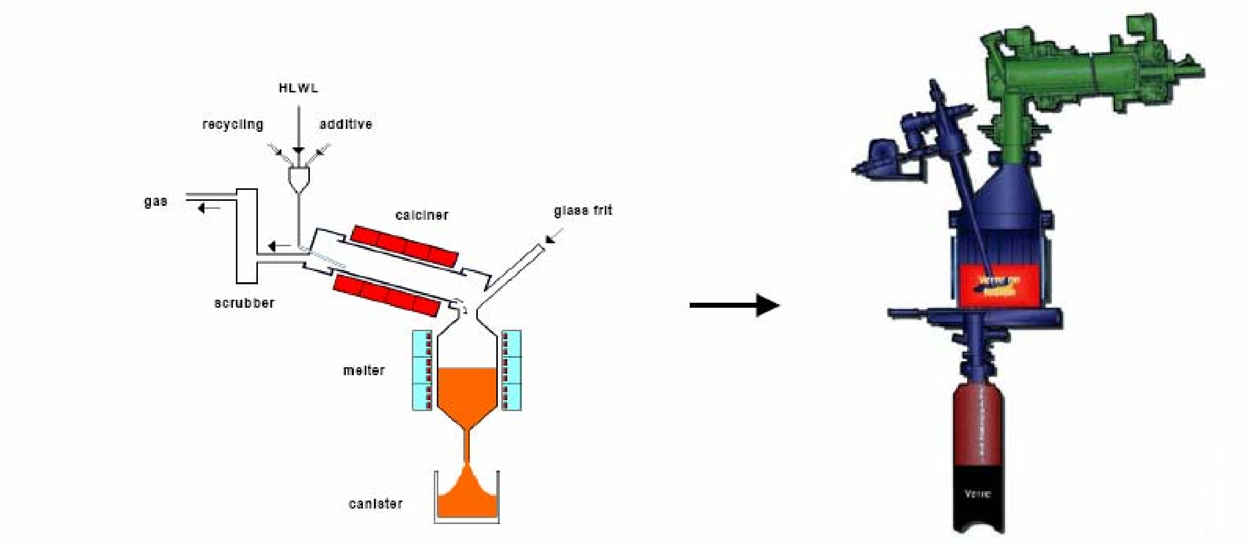


Fig. 1. Two-step vitrification process

The inductor is outside and connected to a high frequency generator, thus:

- Independent from the melting pot,
- Not sensitive to the glass melting (no wear, no corrosion, no shorting),
- Not directly contaminated by High Active Level glass
- Easy to start (even if the metallic melter is full or empty), to stop, to maintain or to replace.

As the metallic walls of the crucible are cooled, a thin layer of solidified cold glass is created. This layer, also called 'skull-melter', protects the crucible from the high temperature and from corrosion. As results, the process time life is upgraded and higher temperature of the glass can be reached.

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. Despite the high temperature of the molten glass (1200 to 1280°C), this layer limited the quantity of volatilized material.

Induction technology development on a full-scale prototype was conducted in parallel with glass formulation studies. For example, The UMo glass electrical resistivity complies with its fabrication in a cold crucible melter; the thermal conductivity was optimized to diminish radiated heat loss to the melter walls in compliance with glass formulation to maintain the final quality product.
The way from preliminary R&D choices to large scale industrial implementation 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. 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 BUBBLING

In order to model air bubbling in the molten glass, specific experiments have been performed based on the principle of hydraulic similarity. They consist in replacing the glass by silicon oil of equivalent viscosity in a tank of the same size. Reynolds number as Galilee numbers are well respected between the two configurations. Only the Morton number differs, but it reflects tension surface effect which has been proven to have a weak effect on the formation of air bubbles in a very viscous liquid [6]. Figure 2a shows an experimental visualization of the bubbling in a simplified tank with a unique central bubbler.

These experiments allow us to choose and validate correlations predicting equivalent diameter of the bubble and bubble velocity. These correlations then are used in a so called 'one-mesh' model (*cf.* fig. 2b) to simulate the recirculation flow induced by the bubble stream rising in the glass. This model, due to Snabre *et al.* [6,7], consists in building a zone corresponding to the stream of the bubbles in which a vertical constant volumic force is applied to the fluid. Refers to [7] for the expression of the equations of the model.

It has been implanted in the commercial software Fluent®. A numerical simulation of the time-average flow induced by the bubbles is obtained. This model is quite simple and will be easily integrated in the coupling of thermal and induction phenomena presented in the next section.

Experimental velocity profiles obtained from PIV (Particle Imaging Velocimetry) measurement are available. The average oil flow is directly compared to numerical results on figure 2 (c & d) for five different values of airflow injected: 100, 250, 500, 750 and 1000 NL/h.

The model is still currently in process of optimization and will be added to the real configuration modelling with the coupling of thermal, induction and hydraulic phenomena.

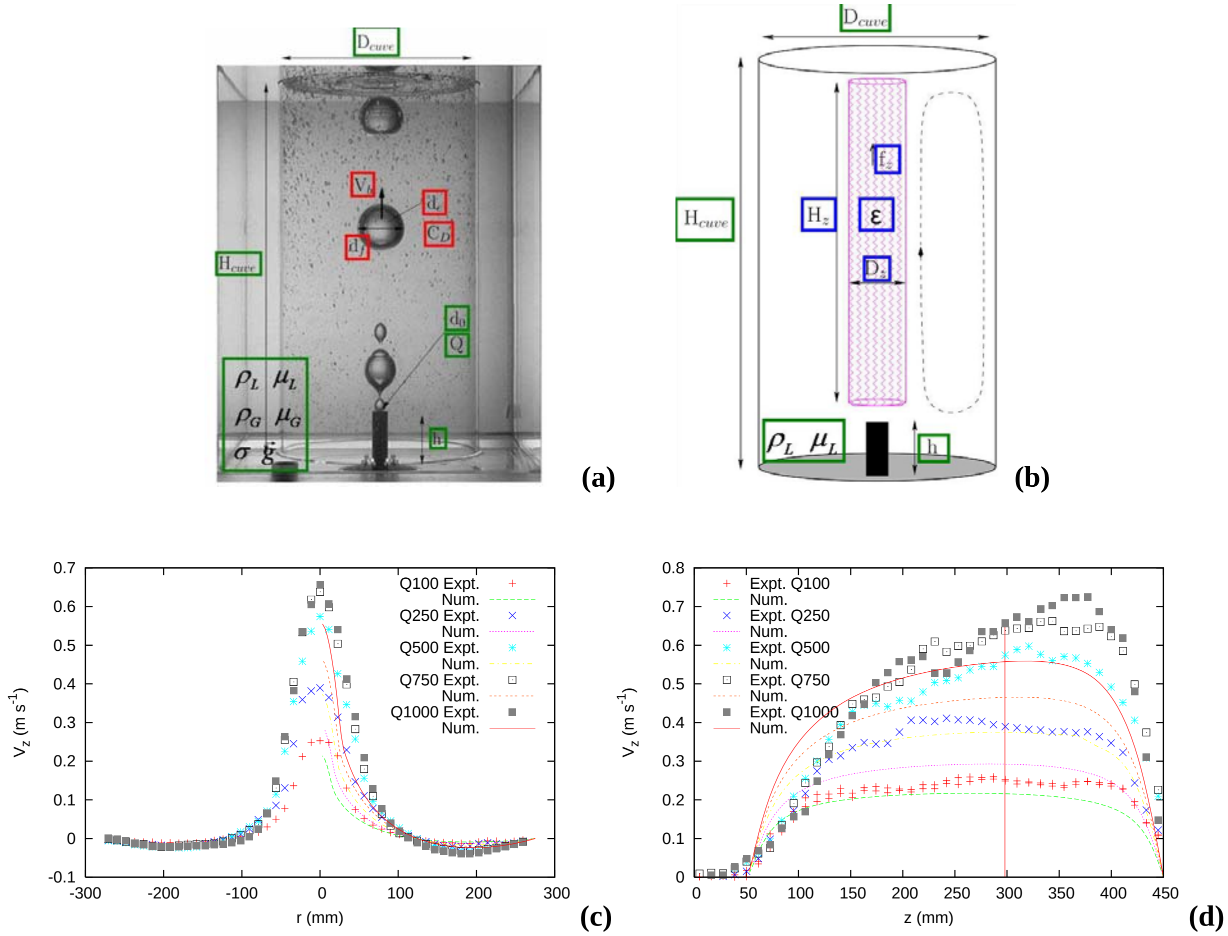


Fig. 2. ***Overview of the bubbling (a), numerical model and comparison of numerical (b) and experimental vertical velocity profile on a radius at z=298mm (c) and along the vertical axis for five value of airflow Q (d)***

MODELLING OF INDUCTION HEATING COUPLED TO THERMO-HYDRODYNAMIC

For this study a coupling between induction, thermal and hydrodynamic equations is built. An application case is shown in which for simplicity the mechanical stirring and bubbling is not modelled. Only thermo convectives forces will acts on the flow.

Results are compared with those of Jacoutot [3,4] obtained with simulation in which induction heating is solved under axisymmetric assumption. This former coupling will be called 'pseudo-3D coupling'.

Principles

This study is a continuation of previous works modelling this process (Jacoutot [3,4]). 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 software’s is achieved. Fluent® is used to solve hydrodynamic and thermal equations whereas Flux® computes the Maxwell equations. The electrical conductivity and the density of the Joule power are used to couple electromagnetism and thermo-hydrodynamics. All computations are three-dimensional. The coupling is based on files data transfers between the two software’s.

Geometry

In this study, we consider a molten glass of 185 mm of high, with 250 mm of radius. The mechanical stirrer is not modelled, only four thermocouples within a sheath made of alumina (electric insulated) are taken into account.

Electromagnetic model

The commercial software Flux® is used to solve induction equations. The crucible as well the inductor are not modelled, 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.

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

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.

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

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

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

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

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:

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

The convection is driven by two phenomena. First, buoyancy forces are modelled in the Boussinesq approximation (last term of right hand side of equation (2)). Second, thermo capillary 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: $\sigma_s\left(T\right)=\sigma_{s0}-\gamma\left(T-T_0\right)$ where $\gamma=-\partial\sigma_s/\partial T$. For almost all liquid, γ is constant and positive and for the glass its value is $10^{-4}\ N.m^{-1}.K^{-1}$[3]. 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:

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

Thermal boundary conditions are handled via a global exchange coefficient on the crucible wall and bottom. At the free surface a mixed condition convection-radiation is considered with an emissivity of 0.9.

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 [4] shown, this does not change the results. The flow time step for Fluent® is 1s.
Initialization is performed with the results obtained with the pseudo-3D coupling due to Jacoutot [4]. In this coupling, the electromagnetic computation was axisymmetric.
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^{-8}.

RESULTS

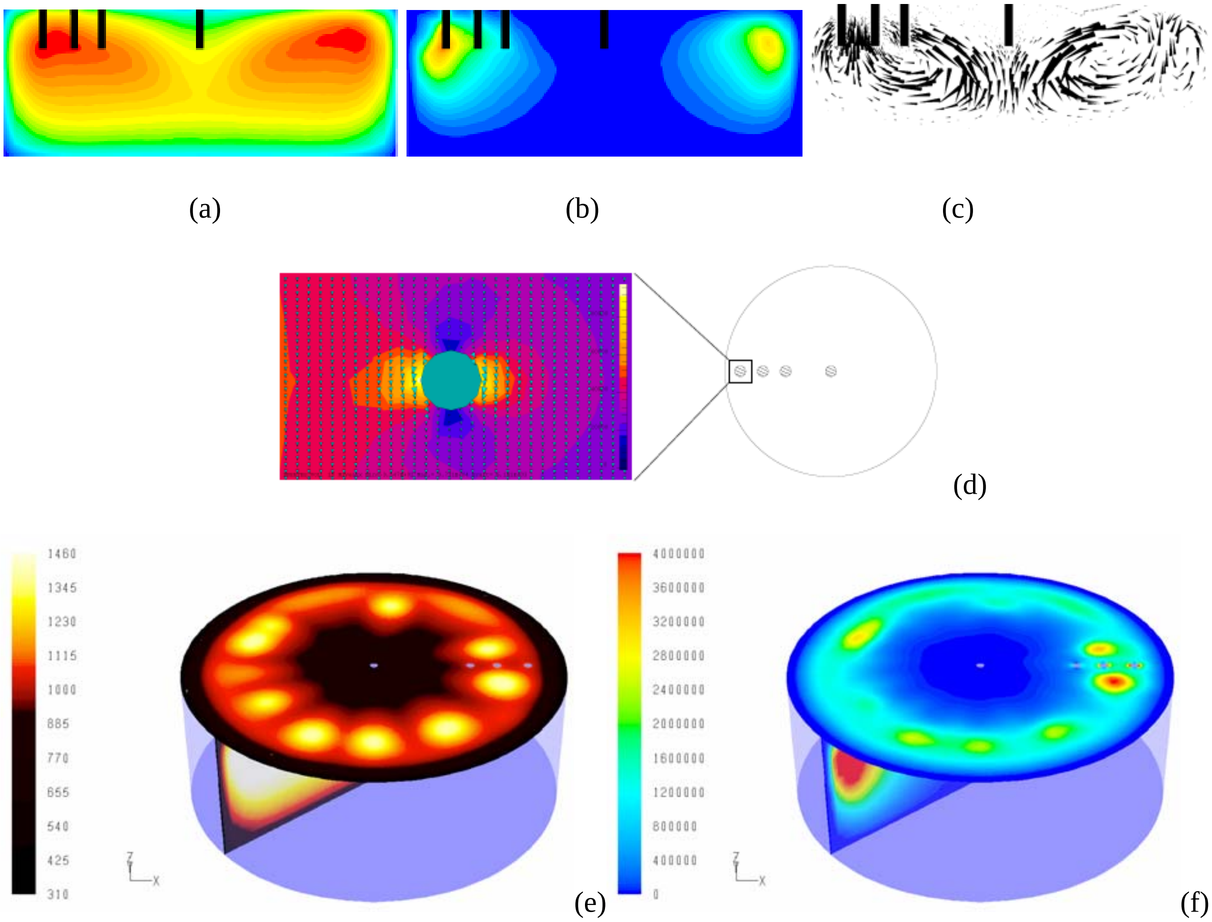
Total Joule power injected of 45 kW

In this first case, a 45kW Joule power heating is imposed in the glass container. Figure 3 (a, b & c) shows iso-contours of temperature and Joule heating in a vertical plane including thermocouples. Cold glass in contact with the cold crucible appears clearly. The maximum Joule power density is located in the hot parts of the molten glass which have a better electrical conductivity. For such a Joule heating power, glass flow due to natural convection and thermo capillarity is stable and stationary. Without thermocouples, an axisymmetric flow appears. Such a result has already been reported by Jacoutot [1]. 3D simulations only exhibit a small perturbation of the Joule heating field near the thermocouples.

Figure 3(d) shows how induced current in the molten glass avoid the sheath of thermocouple made of alumina (electrical insulated). The Joule power then concentrates on both side of the sheath creating two local overheating zones.

Total Joule power injected of 55 kW

For this total Joule power rate, the flow obtained with the pseudo-3D coupling exhibits thermoconvectives circulations [4]. These movements are convection cells of rising hot glass similar to Benard-Marangoni cells [8] (*cf.* figure 3 e & f).



(a) (b) (c)

(d)

(e) (f)

Fig. 3. ***Temperature isocontours (a), Joule power density isocontours (b) and velocity vectors (c) on crossing vertical plane including thermocouples. Scales are respectively [300;1670]K, [0 ;7,76.10^6]W.m^{-3} and [0 ;4,68.10^{-4}] m.s^{-1}; Joule power isocontours and induced current vectors at the free surface near thermocouple TC3 (d) Temperature isocontours (e) and Joule power density isocontours (f) at the free surface and in a crossing vertical plane. Scales are respectively [310;1450]K and [0 ;4.10^6]W.m^{-3}***

When the 3D-3D coupling is performed, the Joule power concentrates in the hotter glass zone increasing temperature differences. Thus, the movement is reinforced, and the convections cell growths and their number decreased. Moreover, this circulation was initially stationary (non time dependent). The figure 4a shows the evolution of the number of convective cells with physical time. The initialization exhibits 22 cells. During the first 1000s, this number decreased to 11 because cells growths, then cells number decrease a second time because of loss of periodicity, and cells appears and disappears in the glass. Cell number oscillates between 5 and 9.

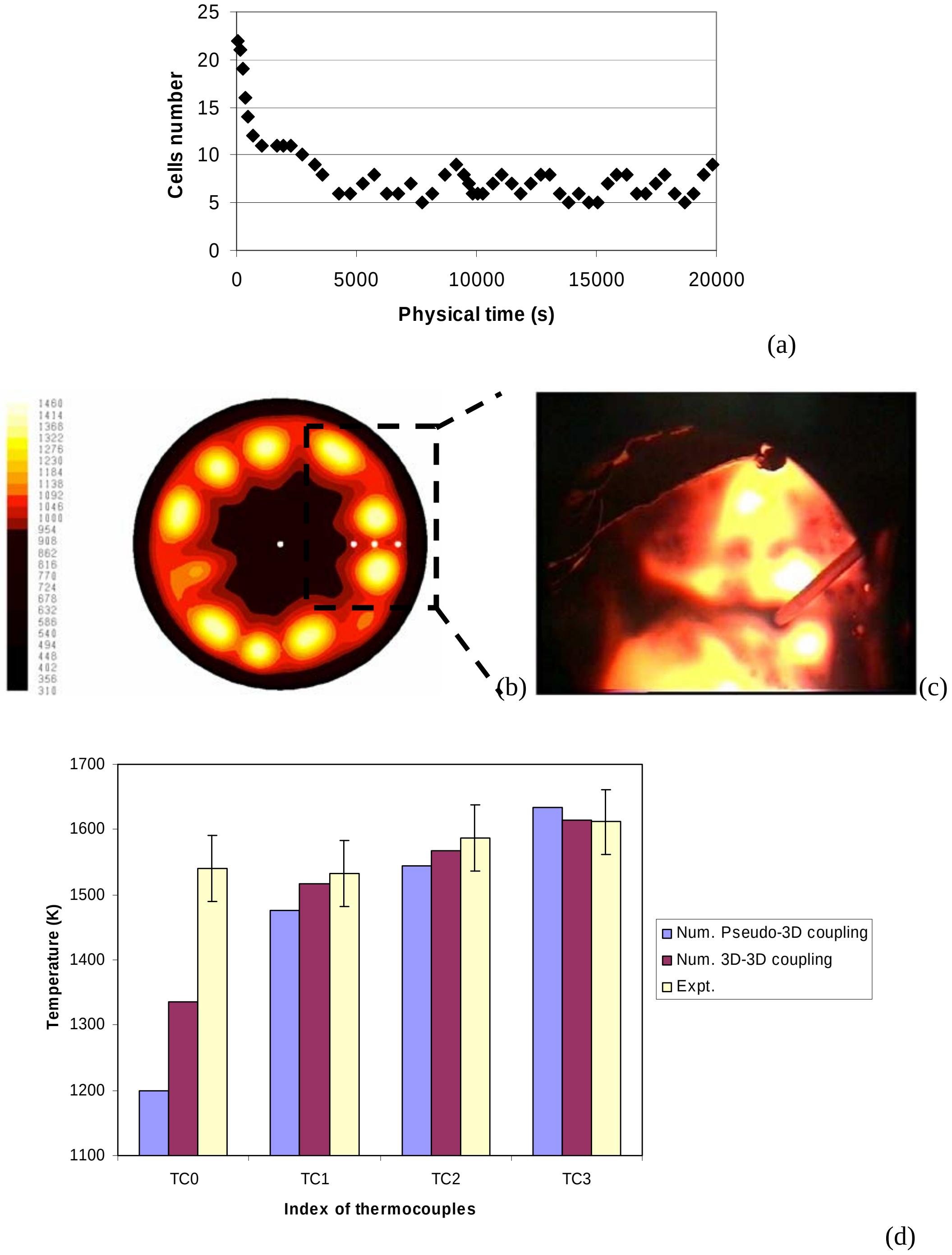


Fig. 4. ***Time evolution of the number of convection cells (a); Numerical (b) and experimental (c) temperature isocontours at the free surface of the molten glass; Comparison of measured temperature and calculated ones at the location of the thermocouples (d).***

Unfortunately, the construction of the classical non-dimensional numbers as Rayleigh or Marangoni is very difficult because physical properties of the glass are strongly dependent of the temperature.

Characteristic time of evolution of convective cell is about ~15min which is in qualitative agreement with experimental visualization. Number of cells in the glass could not be determined experimentally because of the limited view angle of the camera, but size of cells (~100 mm) is in agreement (*cf.* fig. 4 b& c).
Figure 4d shows temperature measurement with the thermocouples compared with calculated temperature with the pseudo-3D and 3D-3D coupling. Results are in good agreement too.

CONCLUSION

A full 3D strong iterative coupling between induction, thermal and hydrodynamic phenomena has been performed using the two commercial software Flux® and Fluent®. This coupling has been used in the simulation of the flow of molten glass heated by direct induction in a cold crucible. In the studied configuration, the 3D coupling enlightens flow patterns, which may occur in the molten glass heated by induction. It has been demonstrated that three-dimensional effect of induction heating could have a significant impact on the flow configuration. If a certain threshold of the total injected power is reached, then 3D thermo convective cells appear. This pattern becomes unsteady and presents chaotic behavior. The wave numbers and also characteristic times are in good agreement with the experimental data, obtained in CEA Marcoule experiments.
It should be specified that these flow patterns are found in this example only because mechanical stirring and air bubbling are not modelled. 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.
On the other hand, a simple numerical model has been developed to simulate the bubbling effect as a driving force on the glass flow. It has been built and validate on a wide range of experimental data. This model will be soon integrated to the full simulation of the process.

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