

The transport and settling of platinum-group-metal particles in glass melts

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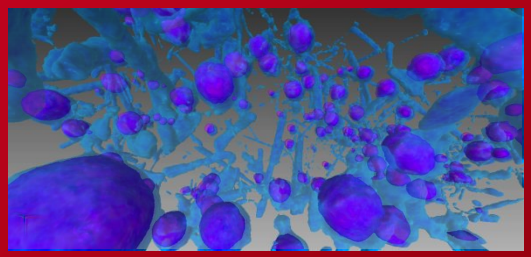
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Society of Glass Technology Centenary Conference
SGT100 & ESG2016
Sheffield UK, September 4th-8th 2016

Waste vitrification

THE TRANSPORT AND SETTLING OF PLATINUM-GROUP-METAL PARTICLES IN GLASS MELTS

PRESENTATION BY GUILLAUME BARBA ROSSA

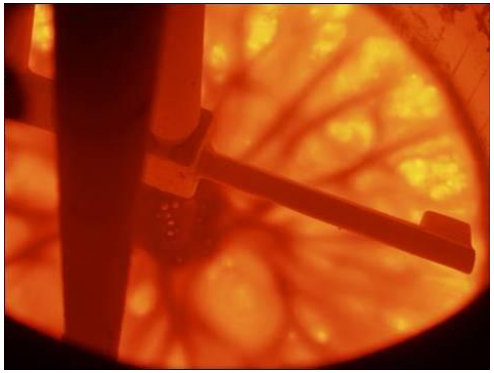
G. BARBA ROSSA, E. SAUVAGE
CEA, DEN, DTCD, SCDV, LDPV
BP 17171, F-30207 BAGNOLS-SUR-CEZE, FRANCE

Short overview of the High-Level-Waste confinement process

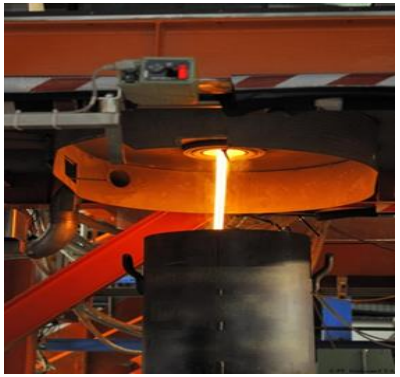
- Ensure long-time confinement of High-Level-Waste (HLW) of spent nuclear fuel reprocessing
- Vitrification: atomic-scale incorporation of HLW in a glass melt
- Most recently developed technology used for vitrification
  - **Induction-heated cold crucible**



Full scale prototype at CEA



Free surface of the melt in the crucible

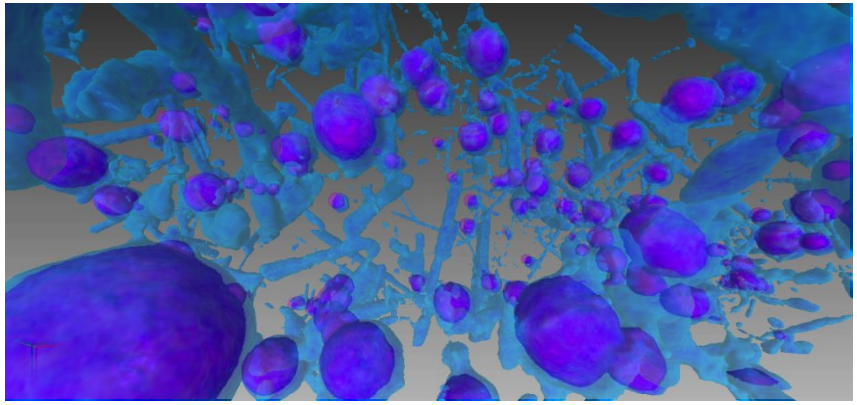


Glass pouring

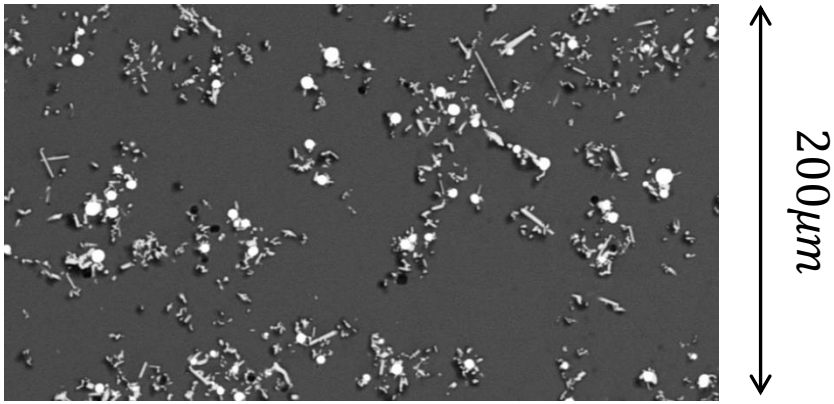
Particles encountered in nuclear waste vitrification processes

- Nuclear glass melt
  - Homogeneous borosilicate glass matrix
  - Seeded with small particles ($\sim 10\mu m$)

- Particles are Platinum-Group-Metals with a **much higher density than glass**
  - Ruthenium dioxide (RuO_2) needles
  - Palladium (Pd) spheres



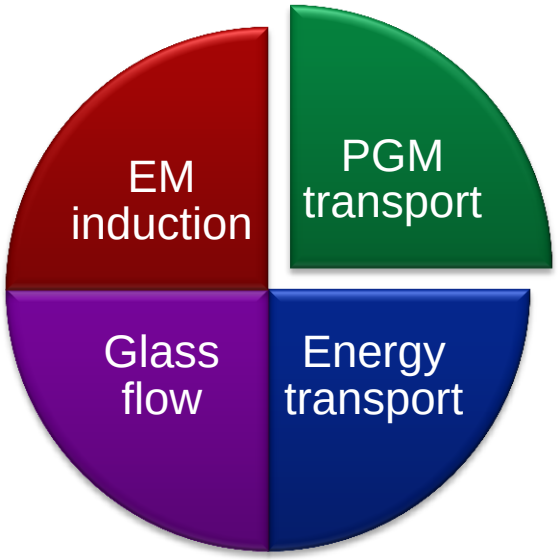
Iso-contours of density from X-ray microtomography (made at ESRF)

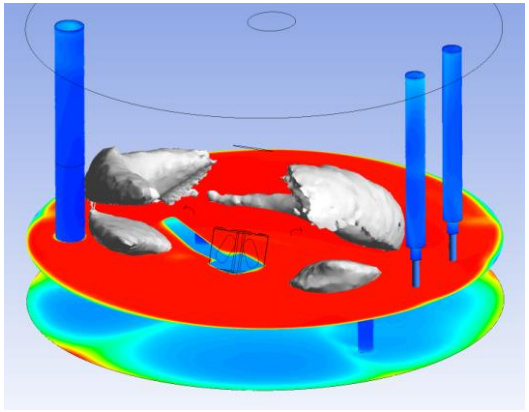


SEM

NOVITOM
ADVANCED 3D MICRO-IMAGING

Numerical simulation of the vitrification process in cold crucibles



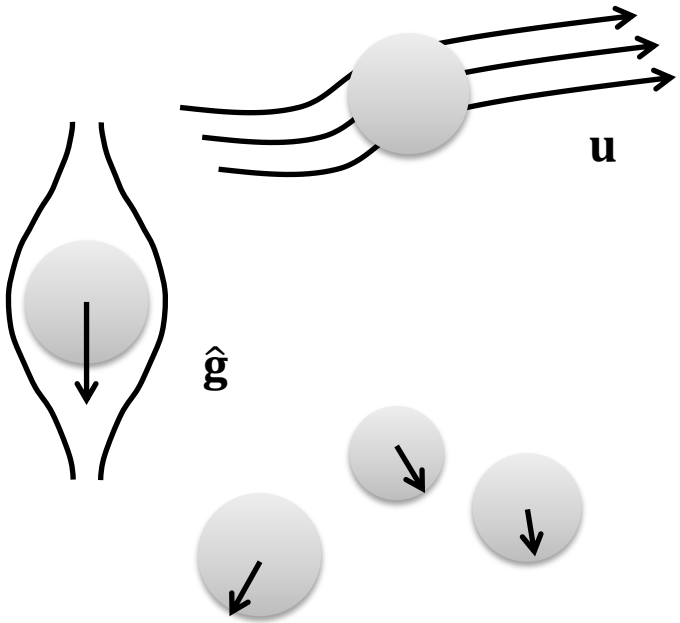


Full thermo-hydraulic numerical simulation of the cold crucible

- Numerical simulation of coupled models
  - Fluid mechanics for stirred glass: Navier-Stokes equations
  - Temperature transport: Energy equation
  - Electromagnetics for induction: Maxwell equations
- But PGM particles have significant effects when their local volume fraction increases (because of settling)
  - Viscosity increase
  - Electrical conductivity increase
  - …
- Need for a supplementary model to compute **particles local volume fraction in the melt $C(x, y, z, t)$**

Transport model for particles

- Transport phenomena included in a **one-fluid model**
  - Advection by the flowing suspending fluid
  - Hindered settling with Stokes velocity $\tau(C,T)$ (Michaels & Bolger formula)
  - Hydrodynamic diffusion $\kappa(T)$



- Resulting in a single unsteady evolution equation for the particles volume fraction C

$$\frac{\partial C}{\partial t} + \mathbf{u} \cdot \nabla C + \nabla \cdot (\tau \hat{\mathbf{g}} C) = \nabla \cdot (\kappa \nabla C)$$

Accounting for temperature-dependent transport properties

- **4 transport parameters**, 2 of them strongly depend on glass viscosity η

Maximum packing fraction C_a evaluated **from particles' shape**	Hindering exponent $\beta = 4.65$ **computed by Richardson & Zaki**
Settling velocity τ evaluated **from Stokes formula**	Diffusivity κ **fitted in laboratory experiments**

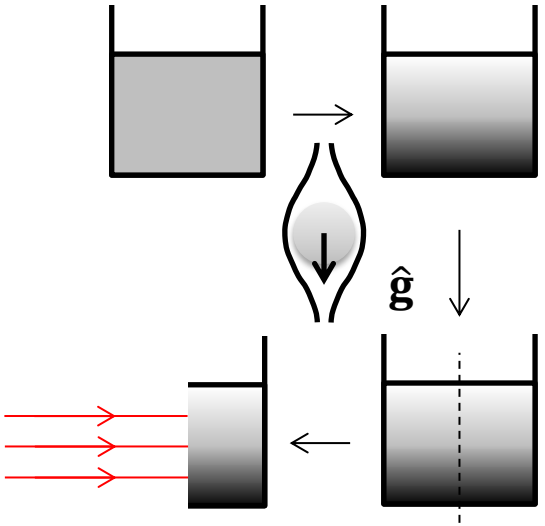
$$\propto \frac{1}{\eta(T)}$$

- Glass melting processes are non-isothermal and glass viscosity depends on temperature

Temperature stratification T → Change in glass viscosity $\eta(T)$ (VFT)

Laboratory settling experiments with a nuclear waste simulant (1)

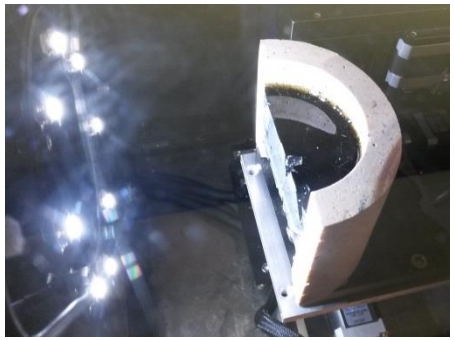
- Initially homogeneous nuclear waste simulant seeded with PGM particles
- Heat-treated during prescribed settling time without any stirring ($\mathbf{u} = 0$)



- Crucible vertically sliced after cooling
- Laser Induced Breakdown Spectroscopy (LIBS) to measure vertical concentration profiles $C(z,t)$

- Comparison to the transport model (solved numerically)

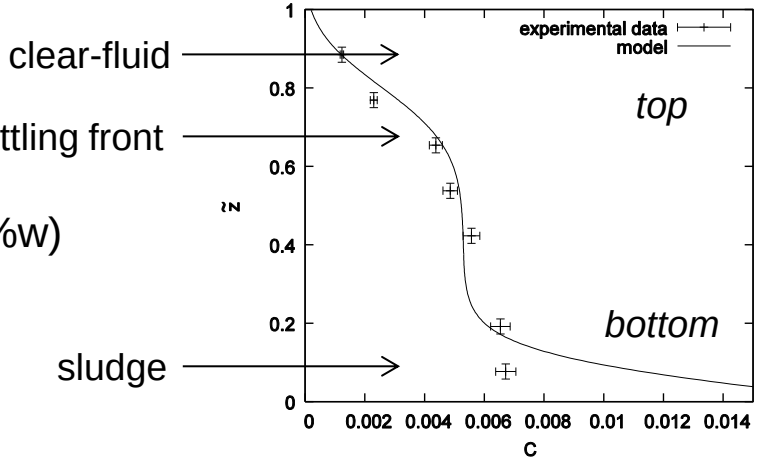
$$\frac{\partial C}{\partial t} + \mathbf{u} \cdot \nabla C + \nabla \cdot (\tau \hat{\mathbf{g}} C) = \nabla \cdot (\kappa \nabla C)$$



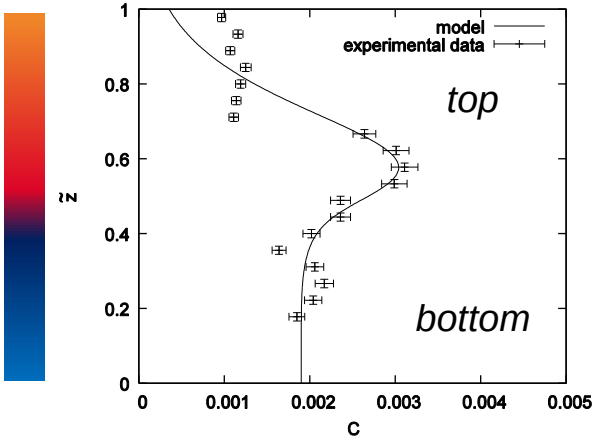
Sliced glass sample in the LIBS apparatus

Laboratory settling experiments with a nuclear waste simulant (2)

- Exp. 1: isothermal case
  - Sample height 2.6cm
  - Uniform temperature 1300°C
  - Ruthenium dioxide particles 0.53%vol (1.5%w)
  - Heating time 16h



- Exp. 2: non-isothermal case
  - Sample height 4.5cm
  - Imposed temperature gradient 592°C-1215°C
  - Palladium particles 0.19%vol (0.9%w)
  - Heating time 65h



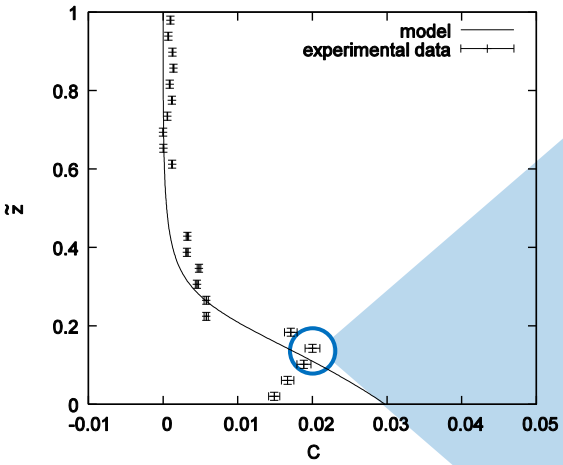
Transport parameters used in simulations (at 1300°C)

Experiment	Particles	$\tau\ (10^{-7} m/s)$	$\kappa\ (10^{-10} m^2/s)$	C_a	β
1	RuO_2	1.3	1.5	8%	4.65
2	Pd	2.9	18	60%	4.65

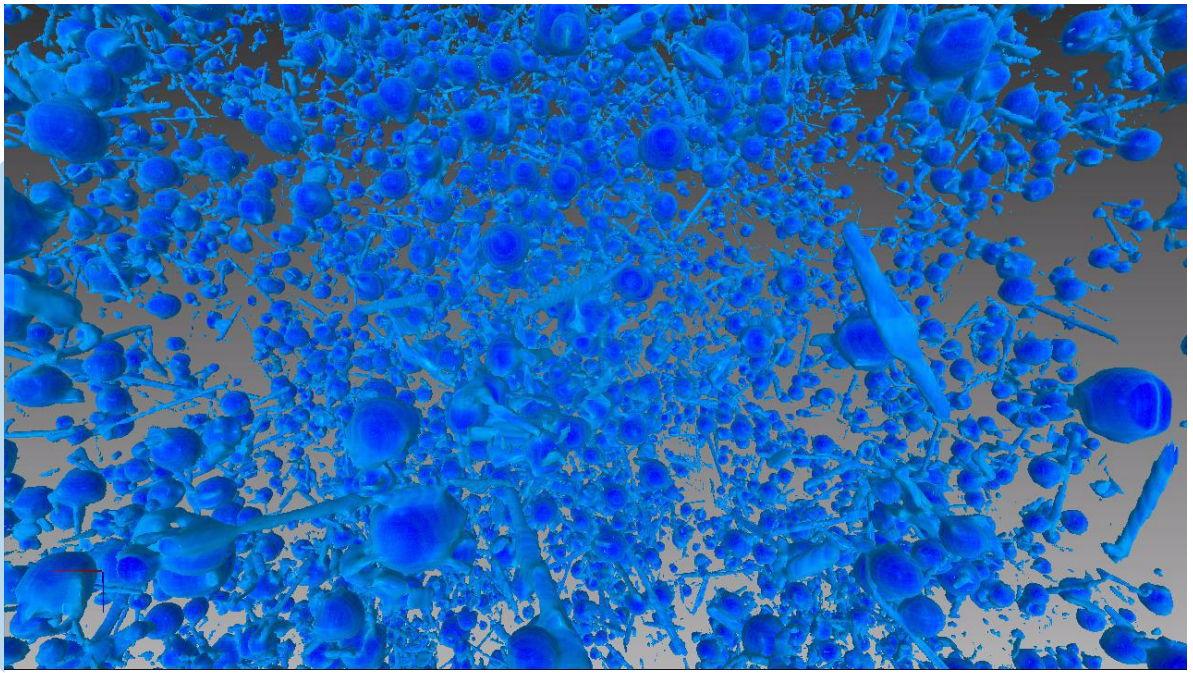
Structure of the sludge

- Exp. 3: sludge obtained from long-term settling
  - Sample height 4.9cm
  - Uniform temperature 1200°C
  - Both ruthenium dioxide and palladium particles 0.7%vol
  - Heating time 211h

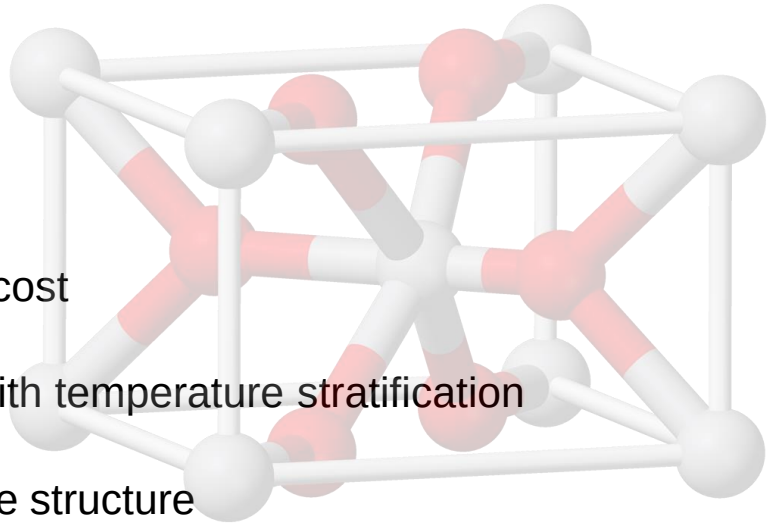
- **Intricate percolated network**
- Relatively low volume fraction 3.5%vol



RuO_2 *volume fraction*



Conclusion



- Accurate transport model with low computational cost
- Able to account for PGM settling in glass melts, with temperature stratification
- Precise 3d imaging gives an insight into the sludge structure

- Transport model included in 3d simulations of waste vitrification in cold crucibles along with:
  - “Cloud settling” through Rayleigh-Taylor instabilities
  - Percolation model for the electrical conductivity of PGM-laden glass

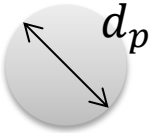
THANK YOU FOR YOUR ATTENTION

Detailed transport model

Hindering function

$$\frac{\partial C}{\partial t} + \mathbf{u} \cdot \nabla C + \nabla \cdot \left(\frac{\tau_0}{f_\eta} \hat{\mathbf{g}} C \overbrace{\left(1 - \frac{C}{C_a} \right)^{\beta}} \right) = \nabla \cdot \left(\frac{\kappa_0}{f_\eta} \nabla C \right)$$

Stokes terminal velocity $\tau_0 = \frac{d_p^2 (\rho_p - \rho_f) g}{18 \eta_0}$



Diffusivity $\kappa_0 = \frac{d_p^2 \alpha}{18 \eta_0}$

Vogel-Fulcher-Tamman viscosity law $\mathrm{f}_\eta \propto \exp\left(\frac{B}{R(T - T_v)}\right)$