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

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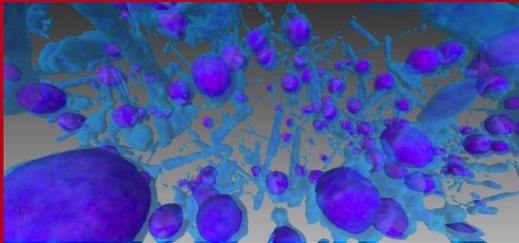
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Society of Glass Technology Centenary Conference

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

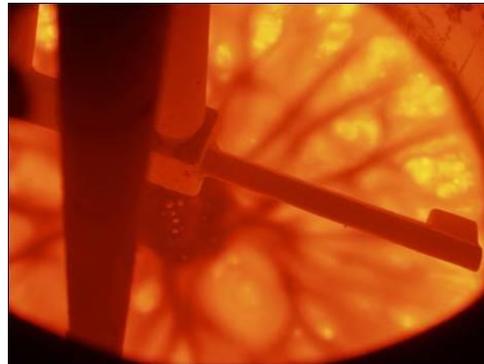
BP 17171, F-30207 BAGNOLS-SUR-CEZE, FRANCE

Short overview of the High-Level-Waste confinement process

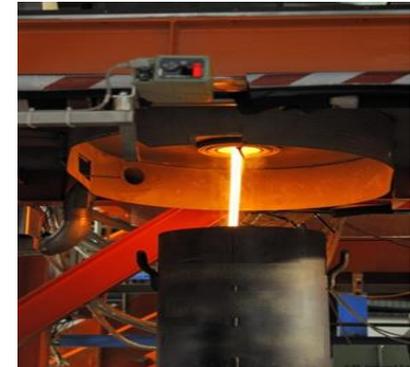
- Ensure long-time confinement of High-Level-Waste (HLW) of spent nuclear fuel reprocessing
- Vitrification: atomic-scale incorporation of HLW in a glass melt
- Most recently developed technology used for vitrification
 - **Induction-heated cold crucible**



Full scale prototype at CEA



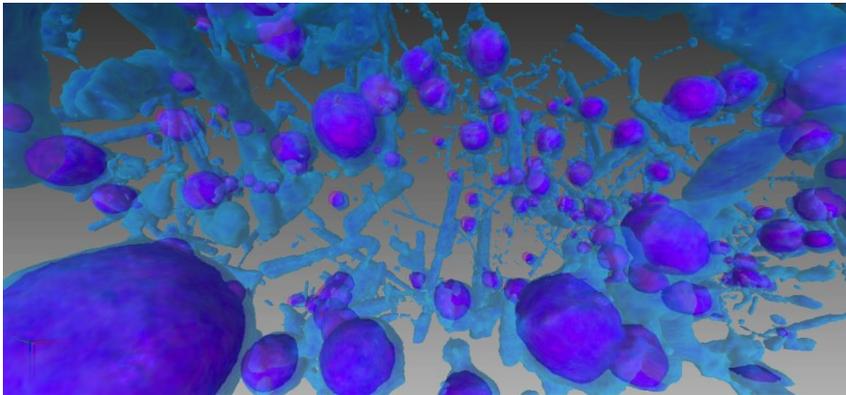
Free surface of the melt in the crucible



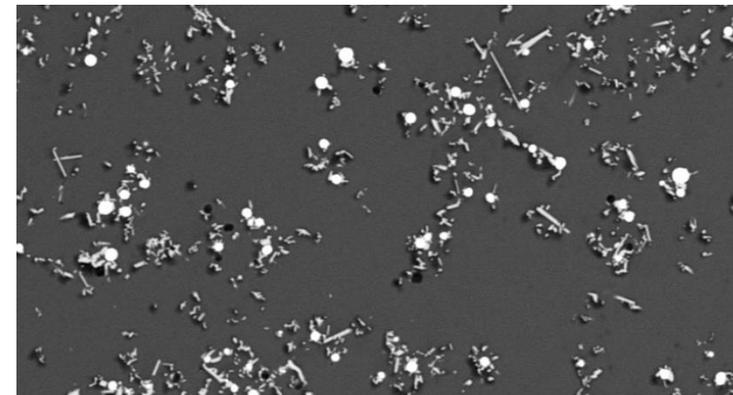
Glass pouring

Particles encountered in nuclear waste vitrification processes

- Nuclear glass melt
 - Homogeneous borosilicate glass matrix
 - Seeded with small particles ($\sim 10\mu\text{m}$)
- Particles are Platinum-Group-Metals with a **much higher density than glass**
 - Ruthenium dioxide (RuO_2) needles
 - Palladium (Pd) spheres



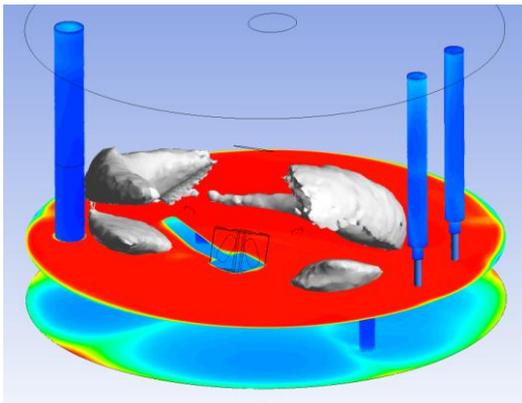
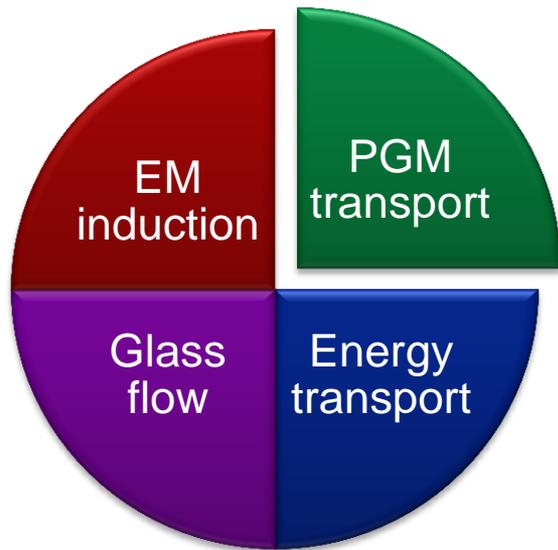
Iso-contours of density from X-ray microtomography (made at ESRF)



SEM

200 μm

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** $C(x, y, z, t)$

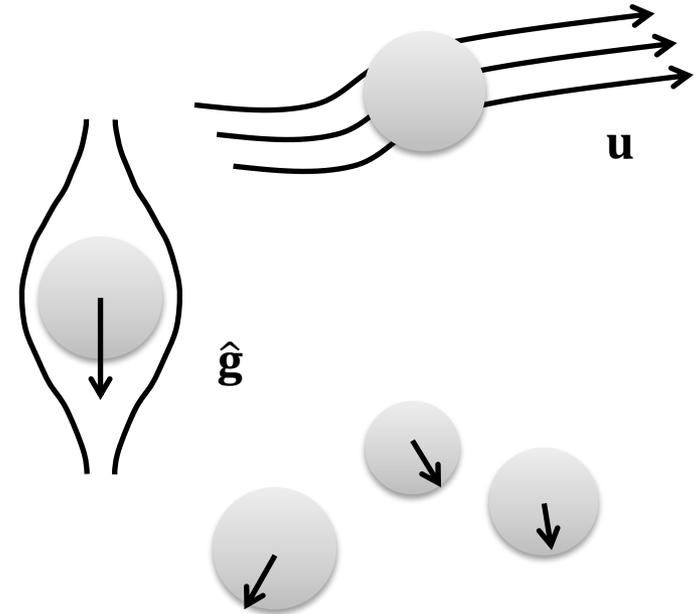
Transport model for particles

■ 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 $\kappa(T)$

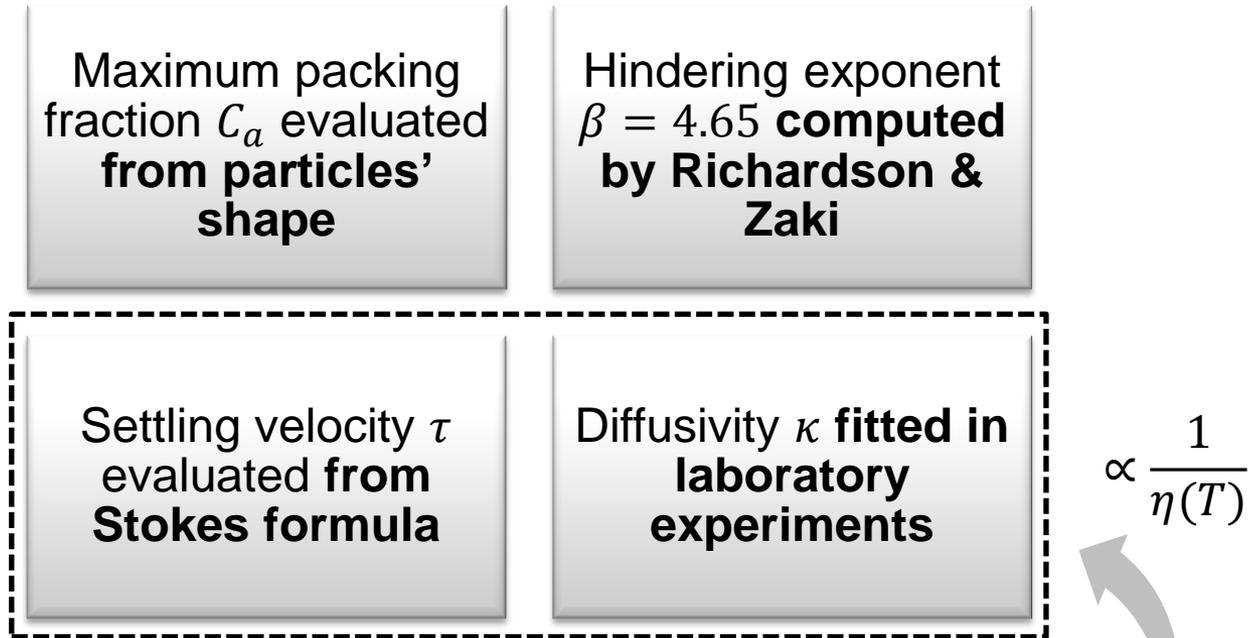


■ Resulting in a single unsteady evolution equation for the particles volume fraction C

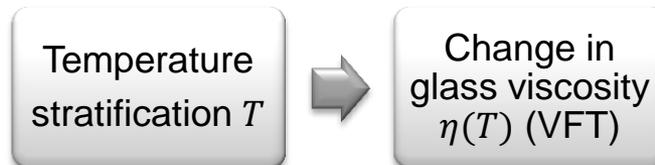
$$\frac{\partial C}{\partial t} + \mathbf{u} \cdot \nabla C + \nabla \cdot (\tau \hat{\mathbf{g}} C) = \nabla \cdot (\kappa \nabla C)$$

Accounting for temperature-dependent transport properties

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



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



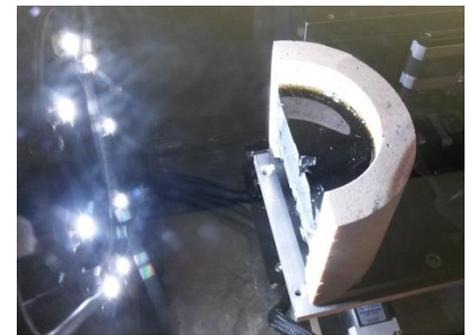
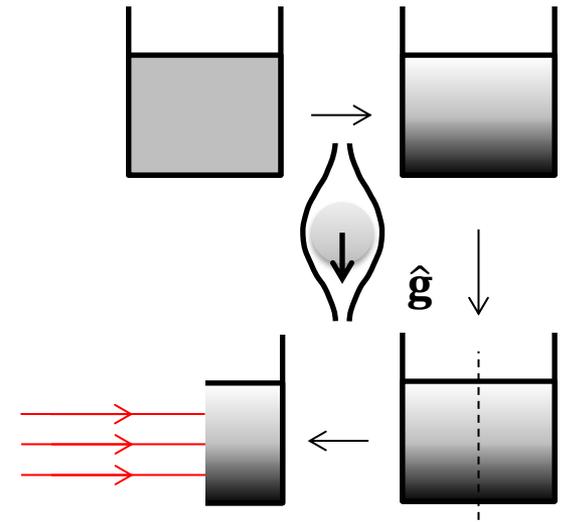
Laboratory settling experiments with a nuclear waste simulant (1)

- 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

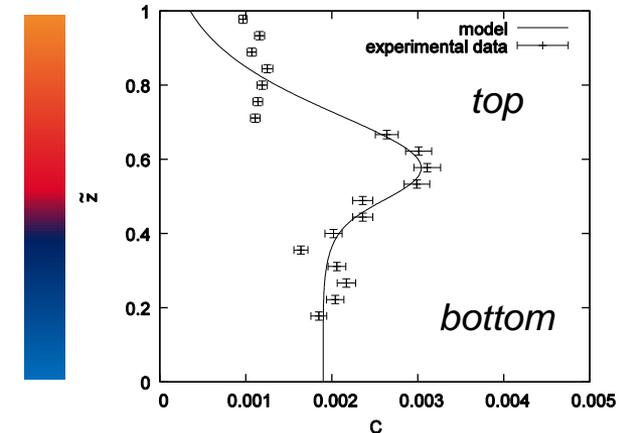
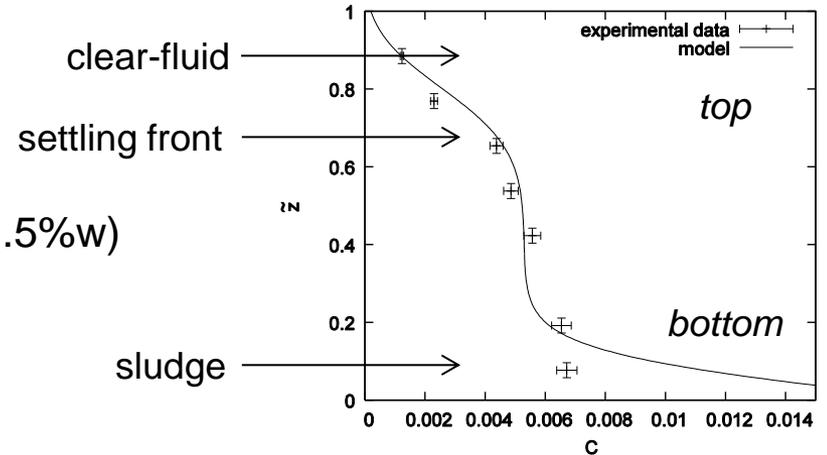
Laboratory settling experiments with a nuclear waste simulant (2)

■ 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



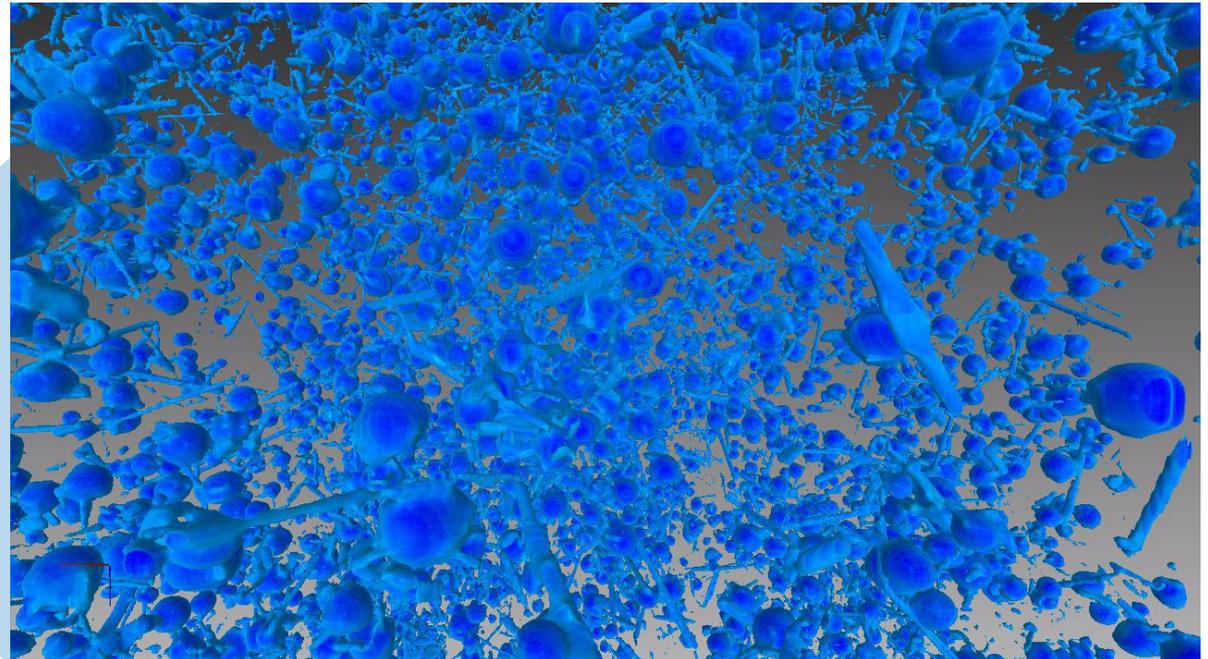
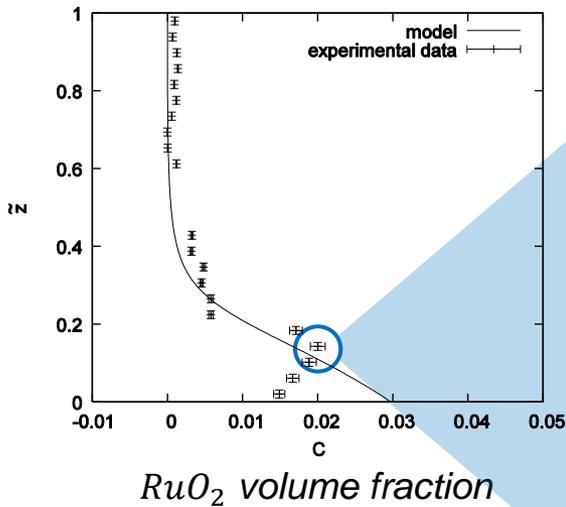
Transport parameters used in simulations (at 1300°C)

Experiment	Particles	τ ($10^{-7}m/s$)	κ ($10^{-10}m^2/s$)	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

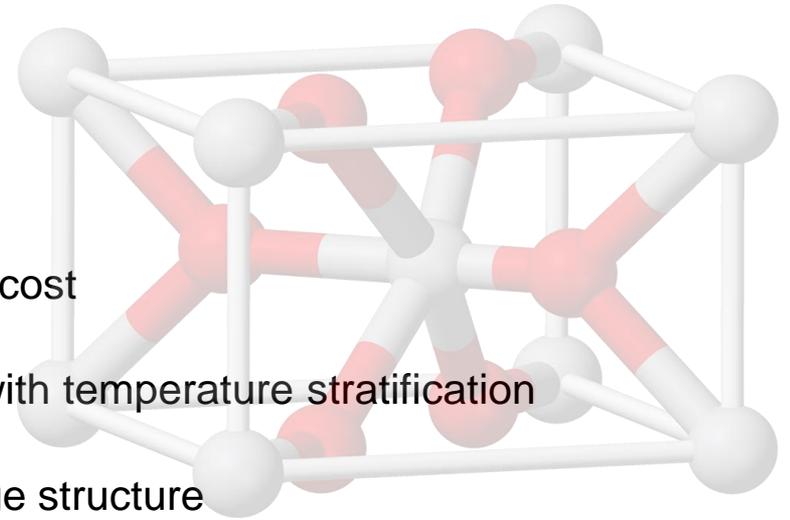
- **Intricate percolated network**
- Relatively low volume fraction 3.5%vol



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

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

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 $f_\eta \propto \exp\left(\frac{B}{R(T - T_v)}\right)$

